

DECLARATION

I, Koichi OISHI Patent Attorney, of OISHI & PARTNERS, 1-10, Kandasudacho, Chiyoda-ku, Tokyo, Japan, hereby certify that I am the translator of the documents in respect of PCT International Application No. PCT/JP2005/004607 filed on March 16, 2005 and that the following is a true and accurate translation to the best of my knowledge and belief.



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[NAME OF DOCUMENT] SPECIFICATION

[TITLE OF THE INVENTION]

DIELECTRIC PASTE FOR A MULTI-LAYERED CERAMIC
ELECTRONIC COMPONENT AND A METHOD FOR
5 MANUFACTURING A MULTI-LAYERED UNIT FOR A
MULTI-LAYERED CERAMIC ELECTRONIC COMPONENT

[FIELD OF THE INVENTION]

[0001]

10 The present invention relates to a dielectric paste for a spacer layer of a multi-layered ceramic electronic component and a method for fabricating a multi-layered unit for a multi-layered ceramic electronic component, and particularly to a dielectric paste for a spacer layer of a multi-layered ceramic electronic component which does not dissolve a
15 binder contained in a layer adjacent to the spacer layer of the multi-layered ceramic electronic component and can reliably prevent defects from being generated in a multi-layered ceramic electronic component and a method for fabricating a multi-layered unit for a multi-layered ceramic electronic component.

20

[BACKGROUND OF THE INVENTION]

[0002]

Recently, the need to downsize various electronic devices makes it necessary to downsize the electronic components incorporated in the devices and improve the performance thereof. Also in multi-layered ceramic electronic components, such as multi-layered ceramic capacitors, it is strongly required to increase the number of layers and make the laminated unit thinner.

[0003]

When a multi-layered ceramic electronic component as typified by a multi-layered ceramic capacitor is to be manufactured, ceramic powders, a binder such as an acrylic system resin, a butyral resin or the like, a 5 plasticizing agent such as a phthalate ester, glycol, adipate ester, phosphate ester or the like, and an organic solvent such as toluene, methyl ethyl ketone, acetone or the like are mixed and dispersed, thereby preparing a dielectric paste for a ceramic green sheet.

[0004]

10 The dielectric paste is then applied onto a support sheet made of polyethylene terephthalate (PET), polypropylene (PP) or the like using an extrusion coater, a gravure coater or the like to form a coating layer and the coating layer is heated to dryness, thereby fabricating a ceramic green sheet.

15 [0005]

Further, a conductive powder of nickel or the like and a binder are dissolved into a solvent such as terpineol, thereby preparing a conductive paste and the thus prepared conductive paste is printed on the ceramic green sheet in a predetermined pattern using a screen printing machine 20 and dried, thereby forming an electrode layer.

[0006]

When the electrode layer has been formed, the ceramic green sheet on which the electrode layer is formed is peeled off from the support sheet to form a multi-layered unit including the ceramic green sheet and the 25 electrode layer. Then, a ceramic green chip is formed by laminating a desired number of the multi-layered units to form the laminated body, pressing the laminated body and dicing the laminated body.

[0007]

Finally, the binder is removed from the green chip, the green chip is baked and an external electrode is formed, thereby completing a multi-layered ceramic electronic component such as a multi-layered ceramic capacitor.

5 [0008]

At present, the need to downsize electronic components and improve the performance thereof makes it necessary to set the thickness of the ceramic green sheet determining the spacing between layers of a multi-layered ceramic capacitor to be equal to or smaller than 3 μm or 2 10 μm and to laminate three hundred or more multi-layered units each including a ceramic green sheet and an electrode layer.

[0009]

However, in a conventional multi-layered ceramic capacitor, since an electrode layer is formed on the ceramic green sheet in a 15 predetermined pattern, a step is formed between the surface of the electrode layer and the surface of the ceramic green sheet where no electrode layer is formed. Therefore, in the case of laminating a number of multi-layered units each including a ceramic green sheet and an electrode layer, it is difficult to bond the ceramic green sheets included in the 20 number of multi-layered units in a desired manner so that the laminated body fabricated by laminating the number of multi-layered units is often deformed and delamination of layers sometimes occurs.

[0010]

In order to solve these problems, it has been proposed to eliminate 25 steps on the surface of the ceramic green sheet by printing a dielectric paste on the surface of the ceramic green sheet in a complementary pattern to that of the electrode layer, thereby forming a spacer layer between neighboring electrode layers.

[0011]

In the case where the spacer layer is printed on the ceramic green sheet between neighboring electrode layers in this manner, thereby fabricating the multi-layered unit, steps on the surface of the ceramic green sheet of each multi-layered unit can be eliminated and even in the case of laminating a number of multi-layered units each including a ceramic green sheet and an electrode layers and fabricating a multi-layered ceramic capacitor, it is possible to bond the ceramic green sheets included in the number of multi-layered units in a desired manner and it is possible to prevent the laminated body fabricated by laminating a number of multi-layered units each including the ceramic green sheet and the electrode layer from being deformed.

[DISCLOSURE OF THE INVENTION]

15 [PROBLEMS TO BE SOLVED BY THE INVENTION]

[0012]

However, in the case where a spacer layer is formed by printing a dielectric paste prepared using terpineol, which is highly popular as a solvent for a dielectric paste, on a ceramic green sheet formed using a butyral system resin, which is the most popular binder for a ceramic green sheet, the binder contained in the ceramic green sheet is dissolved by terpineol contained in the dielectric paste and the ceramic green sheet is swollen or partially dissolved, whereby voids are generated at the interface between the ceramic green sheet and the spacer layer or fissures or wrinkles are generated on the surface of the spacer layer. As a result, in the case where a multi-layered ceramic capacitor is fabricated by laminating a number of multi-layered units to fabricate a laminated body and baking the laminated body, voids are generated in the multi-layered

ceramic capacitor. Further, in the case where fissures or wrinkles are generated on the surface of the spacer layer, since the portions of the spacer layer where fissures or wrinkles are generated tend to drop off, when a number of multi-layered units are laminated to fabricate a 5 laminated body, the portions of the spacer layer where fissures or wrinkles are generated mix into the laminated body as a foreign substance, thereby causing internal defects in the multi-layered ceramic capacitor and generating voids at portions where the spacer layer is missing.

10 [0013]

One proposed solution for these problems is to employ a hydrocarbon system solvent such as kerosene, decane or the like as the solvent. However, since a hydrocarbon system solvent such as kerosene, decane or the like does not dissolve the binder component used for the 15 dielectric paste, it is impossible to completely replace the conventional solvent such as terpineol with a hydrocarbon system solvent such as kerosene, decane or the like. Therefore, since the acrylic system resin contained in the ceramic green sheet as a binder is still soluble in the solvent contained in the dielectric paste to some extent, it is difficult to 20 prevent generation of pinholes and cracks in the ceramic green sheet in the case where the ceramic green sheet is very thin, and since the viscosity of a hydrocarbon system solvent such as kerosene, decane or the like is lower than that of terpineol, it is difficult to control the viscosity of the conductive paste.

25 [0014]

Further, Japanese Patent Application Laid Open No. 5-325633, Japanese Patent Application Laid Open No. 7-21833 and Japanese Patent Application Laid Open No. 7-21832 propose use of a hydrogenated

terpineol such as dihydroterpineol or a terpene system solvent such as dihydroterpineol acetate instead of terpineol as a solvent. However, since the acrylic system resin contained in the ceramic green sheet as a binder is also soluble in a hydrogenated terpineol such as dihydroterpinyol or a 5 terpene system solvent such as dihydroterpinyol acetate to some extent, it is difficult to prevent generation of pinholes and cracks in a ceramic green sheet in the case where the ceramic green sheet is very thin.

[0015]

It is therefore an object of the present invention to provide a 10 dielectric paste for a spacer layer of a multi-layered ceramic electronic component which does not dissolve a binder contained in a layer adjacent to the spacer layer of the multi-layered ceramic electronic component and can reliably prevent defects from being generated in a multi-layered ceramic electronic component.

15 [0016]

Another object of the present invention is to provide a method for 20 fabricating a multi-layered unit for a multi-layered ceramic electronic component which can reliably prevent defects from being generated in a multi-layered ceramic electronic component and form a spacer layer in a desired manner.

[MEANS FOR SOLVING THE PROBLEMS]

[0017]

The inventors of the present invention vigorously pursued a study 25 for accomplishing the above objects and, as a result, made the discovery that in the case where a dielectric paste for forming a spacer layer was prepared using ethyl cellulose having an apparent weight average molecular weight of 110,000 to 190,000 as a binder and at least one kind

of solvent selected from the group consisting of isobornyl acetate, dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate, it was possible to prepare a dielectric paste having a 5 viscosity suitable for printing and dissolve the binder of the dielectric paste in the solvent in a desired manner and even when the dielectric paste was printed on a ceramic green sheet, thereby forming a spacer layer, the binder contained in the ceramic green sheet was not dissolved in the solvent contained in the dielectric paste and it was therefore 10 possible to reliably prevent the ceramic green sheet from being swollen or partially dissolved so as to generate voids at the interface between the ceramic green sheet and the spacer layer or generate fissures or wrinkles on the surface of the spacer layer, and it was therefore possible to effectively prevent voids from being generated in a multi-layered ceramic 15 electronic component such as a multi-layered ceramic capacitor.

[0018]

The present invention is based on these findings and therefore, the objects of the present invention can be accomplished by a dielectric paste for a spacer layer containing ethyl cellulose having an apparent weight 20 average molecular weight of 110,000 to 190,000 as a binder and at least one kind of solvent selected from the group consisting of isobornyl acetate, dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate.

25 In the present invention, a dielectric paste for a spacer layer is prepared by kneading a dielectric material (ceramic powder) and an organic vehicle obtained by dissolving ethyl cellulose having an apparent weight average molecular weight of 110,000 to 190,000 into an organic

solvent.

The dielectric material can be selected from among various compounds capable of forming a composite oxide or oxide, such as a carbonate, nitrate, hydroxide, organic metallic compound and the like and mixtures thereof. It is preferable to use a dielectric powder having the same composition as that of a dielectric powder contained in a ceramic green sheet described later. The dielectric material is normally used in the form of a powder whose average particle diameter is about 0.1 μm to about 3.0 μm .

In the present invention, it is preferable for the dielectric paste to contain ethyl cellulose having an apparent weight average molecular weight of 115,000 to 180,000.

[0019]

In the present invention, it is possible to mix two or more kinds of ethyl cellulose having different average molecular weights so as to adjust an apparent weight average molecular of the ethyl cellulose to 110,000 to 190,000 or use ethyl cellulose having a weight average molecular weight of 110,000 to 190,000 so as to adjust an apparent weight average molecular of the ethyl cellulose to 110,000 to 190,000. In the case of adjusting an apparent weight average molecular of the ethyl cellulose by mixing two or more kinds of ethyl cellulose having different average molecular weights, an apparent weight average molecular of the ethyl cellulose can be adjusted to 130,000 to 190,000 by mixing ethyl cellulose having a weight average molecular weight of 75,000 and ethyl cellulose having a weight average molecular weight of 130,000 or mixing ethyl cellulose having a weight average molecular weight of 130,000 and ethyl cellulose having a weight average molecular weight of 230,000, for example.

The dielectric paste for forming a spacer layer preferably contains about 4 weight parts to about 15 weight parts, more preferably, about 4 weight parts to about 10 weight parts of ethyl cellulose and preferably contains 40 weight parts to about 250 weight parts, more preferably, 60 weight parts to about 140 weight parts, most preferably, 70 weight parts to about 120 weight parts of a solvent with respect to 100 weight parts of a powder of a dielectric raw material.

The dielectric paste for forming a spacer layer contains, in addition to the powder of a dielectric raw material and the ethyl cellulose.

The plasticizing agent contained in the dielectric paste for forming a spacer layer is not particularly limited and illustrative examples thereof include phthalate ester, adipic acid, phosphate ester, glycols and the like. The plasticizing agent contained in the dielectric paste for forming a spacer layer may or may not belong to the same plasticizing agent group as that of a plasticizing agent contained in a ceramic green sheet described later. The dielectric paste for forming a spacer layer contains the plasticizing agent in an amount of about 0 weight part to about 200 weight parts with respect to 100 weight parts of the ethyl cellulose, preferably in an amount of about 10 weight parts to about 100 weight parts, most preferably in an amount of about 20 weight parts to about 70 weight parts.

The release agent contained in the dielectric paste for forming a spacer layer is not particularly limited and illustrative examples thereof include paraffin, wax, silicone oil and the like. The dielectric paste for forming a spacer layer contains the releasing agent preferably in an amount of about 0 weight % to about 100 weight % with respect to 100 weight parts of the ethyl cellulose, preferably in an amount of about 2 weight parts to about 50 weight parts, more preferably in an amount of

about 5 weight parts to about 20 weight parts.

[0020]

The above object of the present invention can be also accomplished by a method for fabricating a multi-layered unit for a multi-layered 5 ceramic electronic component comprising a step of printing a dielectric paste for a spacer layer containing ethyl cellulose having an apparent weight average molecular weight of 110,000 to 190,000 as a binder and at least one kind of solvent selected from the group consisting of isobornyl acetate, dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl 10 acetate, β -dihydrocarvyl acetate, β -menthyl acetate, β -menthone, β -perillyl acetate and β -carvyl acetate on a ceramic green sheet containing an acrylic system resin as a binder in a predetermined pattern, thereby forming a spacer layer.

[0021]

15 According to the present invention, it is possible to prepare a dielectric paste having a viscosity suitable for printing and form a spacer layer in a desired manner. Further, according to the present invention, even when the dielectric paste is printed on a very thin ceramic green sheet containing a butyral system resin as a binder to form a spacer layer, 20 since the binder contained in the ceramic green sheet is not dissolved in the solvent contained in the dielectric paste, it is possible to reliably prevent the ceramic green sheet from being swollen or partially dissolved so as to generate voids at the interface between the ceramic green sheet and the spacer layer or generate fissures or wrinkles on the surface of the 25 spacer layer, and it is therefore possible to reliably prevent voids from being generated in a multi-layered ceramic electronic component such as a multi-layered ceramic capacitor.

[0022]

In the present invention, it is preferable for a dielectric paste to contain ethyl cellulose having an apparent weight average molecular weight of 115,000 to 180,000 as a binder.

[0023]

5 Here, the apparent weight average molecular weight of ethyl cellulose may be adjusted by mixing two or more kinds of ethyl cellulose having different weight average molecular weights so as to be 115,000 to 180,000 or by using ethyl cellulose having a weight average molecular weight of 115,000 to 180,000 so as to be 115,000 to 180,000.

10 [0024]

In the present invention, it is preferable for the degree of polymerization of a butyral system resin contained in a ceramic green sheet as a binder to be equal to or larger than 1000.

[0025]

15 In the present invention, it is preferable for the degree of butyralization of butyral system resin contained in a ceramic green sheet as a binder to be equal to or larger than 64 mol % and equal to or smaller than 78 mol %.

[0026]

20 In a preferred aspect of the present invention, prior to forming the spacer layer or after forming and drying the spacer layer, a conductive paste containing a binder containing ethyl cellulose having a weight average molecular weight of MW_L and ethyl cellulose having a weight average molecular weight of MW_H at a weight ratio of $X : (1-X)$, where
25 MW_L , MW_H and X are selected so that $X * MW_L + (1-X) * MW_H$ falls within a range of 155,000 to 205,000 and at least one solvent selected from the group consisting of isobornyl acetate, dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl acetate, I-dihydrocarvyl acetate,

I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate is printed on a ceramic green sheet in a complementary pattern to that of the spacer layer, thereby forming an electrode layer.

[0027]

5 Since a mixed solvent of terpineol and kerosene, dihydroterpineol, terpineol or like, which is popular as a solvent for an conductive paste for forming an electrode layer, dissolves an acrylic system resin contained in a ceramic green sheet as a binder, when a conductive paste is printed on a ceramic green sheet containing an acrylic system resin as a binder to form
10 an electrode layer, a binder contained in the ceramic green sheet is dissolved by the solvent contained in the conductive paste, whereby pin holes or cracks are generated in the ceramic green sheet. However, according to this preferred aspect of the present invention, since a conductive paste for forming an electrode layer contains a binder
15 containing ethyl cellulose having a weight average molecular weight of MW_L and ethyl cellulose having a weight average molecular weight of MW_H at a weight ratio of $X : (1-X)$, where MW_L , MW_H and X are selected so that $X * MW_L + (1-X) * MW_H$ falls within a range of 155,000 to 205,000 and at least one solvent selected from the group consisting of isobornyl
20 acetate, dihydroterpinal methyl ether, terpinal methyl ether, α -terpinal acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate and the solvent selected from the group consisting of isobornyl acetate, dihydroterpinal methyl ether, terpinal methyl ether, α -terpinal acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate hardly
25 dissolves a butyral system resin contained in a ceramic green sheet as a binder, even when the conductive paste is printed on a very thin ceramic green sheet containing an acrylic system resin to form an electrode layer,

the binder contained in the ceramic green sheet is not dissolved by the solvent contained in the conductive paste and the ceramic green sheet is not swollen or partially dissolved. Therefore, in the case where a ceramic green sheet is very thin, it is reliably possible to prevent pin holes or 5 cracks from being generated in the ceramic green sheet.

[0028]

Moreover, since a conductive paste containing a binder containing ethyl cellulose having a weight average molecular weight of MW_L and ethyl cellulose having a weight average molecular weight of MW_H at a 10 weight ratio of $X : (1-X)$, where MW_L , MW_H and X are selected so that $X * MW_L + (1-X) * MW_H$ falls within a range of 155,000 to 205,000 and at least one solvent selected from the group consisting of isobornyl acetate, dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl acetate, 1-dihydrocarvyl acetate, 1-mentyl acetate, 1-menthone, 1-perillyl acetate 15 and 1-carvyl acetate has a viscosity suitable for printing, an electrode layer can be formed on a ceramic green sheet in a desired manner by printing a conductive paste on the ceramic green sheet in a complimentary pattern to that of a spacer layer.

[0029]

20 Further, in a study done by the inventors of the present invention, it was found that in the case of printing a conductive paste for an electrode layer on a very thin ceramic green sheet to form an electrode layer and printing a dielectric paste for a spacer layer on the very thin ceramic green sheet to form a spacer layer, the solvent contained in the 25 conductive paste for forming the electrode layer and the solvent contained in the dielectric paste for forming the spacer layer dissolved or swelled a binder component contained in the ceramic green sheet and, on the other hand, the conductive paste and the dielectric paste permeated into the

ceramic green sheet, thereby causing short circuit failure and that, therefore, it was preferable to form the electrode layer and the spacer layer on a support sheet separately from the ceramic green sheet and bond it onto the surface of the ceramic green sheet via an adhesive layer 5 after drying it. However, in the case where the electrode layer and the spacer layer are formed on the support sheet separately from the ceramic green sheet in this manner, in order to make the support sheet easy to peel off from the electrode layer and the spacer layer, it is preferable to form a release layer containing the same binder as that contained in the 10 ceramic green sheet on the support sheet and print a conductive paste and a dielectric paste on the release layer, thereby forming an electrode layer and a spacer layer. Even in the case of printing a dielectric paste on the release layer containing the same binder as that contained in the ceramic green sheet to form a spacer layer, when the release layer 15 contains an acrylic system resin as a binder and the dielectric paste contains terpineol as a solvent, the binder contained in the release layer is dissolved by the solvent contained in the dielectric paste so that the release layer is swollen or partially dissolved, whereby voids are generated at the interface between the release layer and the spacer layer 20 or fissures or wrinkles are generated on the surface of the spacer layer. As a result, in the case where a multi-layered ceramic capacitor is fabricated by laminating a number of multi-layered units to fabricate a laminated body and baking the laminated body, voids are generated in the multi-layered ceramic capacitor. Furthermore, in the case where fissures 25 or wrinkles are generated on the surface of the spacer layer, since the portions of the spacer layer where fissures or wrinkles are generated tend drop off, when a number of multi-layered units are laminated to fabricate a laminated body, the portions of the spacer layer where fissures or

wrinkles are generated mix into the laminated body as a foreign substance, thereby causing internal defects in the multi-layered ceramic capacitor and generating voids at portions where the spacer layer were missing.

5 [0030]

However, according to the present invention, since a dielectric paste for forming a spacer layer contains ethyl cellulose having an apparent weight average molecular weight of 110,000 to 190,000 as a binder and at least one kind of solvent selected from the group consisting of isobornyl acetate, dihydroterpinal methyl ether, terpinal methyl ether, α -terpinal acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate and the solvent selected from the group consisting of isobornyl acetate, dihydroterpinal methyl ether, terpinal methyl ether, α -terpinal acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate hardly dissolves a butyral system resin contained in a ceramic green sheet as a binder, even in the case of forming a release layer containing the same binder as that contained in the ceramic green sheet and printing a dielectric paste on the release layer to form a spacer layer, it is possible to reliably prevent the release layer from being swollen or partially dissolved so as to generate voids at the interface between the release layer and the spacer layer or generate fissures or wrinkles on the surface of the spacer layer and it is therefore possible to effectively prevent defects from being generated in a multi-layered ceramic electronic component such as a multi-layered ceramic capacitor.

[TECHNICAL ADVANTAGES OF THE INVENTION]

[0031]

According to the present invention, it is possible to provide a dielectric paste for a spacer layer of a multi-layered ceramic electronic component which does not dissolve a binder contained in a layer adjacent to the spacer layer of the multi-layered ceramic electronic component and 5 can reliably prevent defects from being generated in a multi-layered ceramic electronic component.

[0032]

Further, according to the present invention, it is possible to provide a method for fabricating a multi-layered unit for a multi-layered 10 ceramic electronic component which can reliably prevent defects from being generated in a multi-layered ceramic electronic component and form a spacer layer in a desired manner.

[DESCRIPTION OF THE PREFERRED EMBODIMENTS]

15 [0033]

In a preferred embodiment of the present invention, a dielectric paste for a ceramic green sheet which contains a butyral system resin as a binder is first prepared and is applied onto a long support sheet using an extrusion coater or a wire bar coater, thereby forming a coating layer.

20 A dielectric paste for forming a ceramic green sheet is normally prepared by kneading a dielectric material (ceramic powder) and an organic vehicle obtained by dissolving a butyral system resin into an organic solvent.

[0034]

25 It is preferable for the degree of polymerization of the butyral system resin to be equal to or larger than 1000.

[0035]

Further, it is preferable for the degree of butyralization of butyral

system resin to be equal to or larger than 64 mol % and equal to or smaller than 78 mol %.

An organic solvent used for preparing the organic vehicle is not particularly limited and an organic solvent such as terpineol, butyl 5 carbitol, acetone, toluene, ethyl acetate and the like can be used for preparing the organic vehicle.

The dielectric material can be selected from among various compounds capable of forming a composite oxide or oxide, such as a carbonate, nitrate, hydroxide, organic metallic compound and the like and 10 mixtures thereof. The dielectric material is normally used in the form of a powder whose average particle diameter is about 0.1 μm to about 3.0 μm . The particle diameter of the dielectric raw material is preferably smaller than the thickness of the ceramic green sheet.

The amounts of the respective constituents contained in the 15 dielectric paste is not particularly limited and the dielectric paste may be prepared so as to contain 100 weight parts of a dielectric material, about 2.5 weight part to about 10 weight parts of a butyral system resin and about 50 weight parts to about 300 weight parts of a solvent, for example.

As occasion demands, the dielectric paste may contain additives 20 selected from among various dispersing agents, plasticizing agents, antistatic auxiliary agent, releasing agent, wetting agent and the like. In the case of adding these additives to the dielectric paste, it is preferable to set the total content to be equal to or less than about 20 weight %.

[0036]

As a support sheet coated with the dielectric paste, a polyethylene 25 terephthalate film is employed, for example, and the surface of the support sheet may be coated with a silicon resin, an alkyd resin or the like in order to improve the releasability thereof.

[0037]

The coating layer is then dried at a temperature of about 50 °C to about 100 °C for about 1 to about 20 minutes, whereby a ceramic green sheet is formed on the support sheet.

5 [0038]

In the present invention, the thickness of the ceramic green sheet after drying is preferably equal to or thinner than 3 µm and more preferably equal to or thinner than 1.5 µm.

[0039]

10 Next, a conductive paste for forming an electrode layer is printed on the ceramic green sheet formed on the long support sheet in a predetermined pattern using a screen printing machine, a gravure printing machine or the like.

[0040]

15 It is preferable to form the electrode layer so as to have a dry thickness of about 0.1 µm to about 5 µm and it is more preferable to form the electrode layer so as to have a dry thickness of about 0.1 µm to about 1.5 µm.

20 The conductive paste usable for forming an electrode layer is prepared by kneading a conductive material containing any of various conductive metals or alloys, any of various oxides which will form a conductive material containing any of various conductive metals or alloys after baking, an organic metal compound, resinate or the like, and an organic vehicle prepared by dissolving a butyral system resin in an 25 organic solvent.

[0041]

In this preferred embodiment of the present invention, the conductive paste contains a binder containing ethyl cellulose having a

weight average molecular weight of MW_L and ethyl cellulose having a weight average molecular weight of MW_H at a weight ratio of $X : (1-X)$, where MW_L , MW_H and X are selected so that $X^* MW_L + (1-X)^* MW_H$ falls within a range of 155,000 to 205,000 and at least one solvent selected
5 from the group consisting of isobornyl acetate, dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate.

[0042]

Since the solvent selected from the group consisting of isobornyl acetate, dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate hardly dissolves the butyral system resin contained in a ceramic green sheet as a binder, even in the case of printing the conductive paste on a very thin ceramic green sheet, thereby 15 forming an electrode layer, it is possible to effectively prevent the binder contained in the ceramic green sheet from being dissolved by the solvent contained in the conductive paste, whereby the ceramic green sheet is swollen or partially dissolved. It is therefore possible to reliably prevent generation of pinholes and cracks in the ceramic green sheet even in the
20 case where the ceramic green sheet is very thin.

[0043]

Further, since a conductive paste containing a binder containing ethyl cellulose having a weight average molecular weight of MW_L and ethyl cellulose having a weight average molecular weight of MW_H at a weight ratio of $X : (1-X)$, where MW_L , MW_H and X are selected so that $X^* MW_L + (1-X)^* MW_H$ falls within a range of 155,000 to 205,000 and at least one solvent selected from the group consisting of isobornyl acetate, dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl acetate,
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I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate has a viscosity suitable for printing, it is possible to print a conductive paste on a ceramic green sheet using a screen printing machine, a gravure printing machine or the like to form an electrode layer 5 in a predetermined pattern in a desired manner.

As the conductive material used for preparing the conductive paste, Ni, Ni alloy or the mixture thereof is preferably used. The shape of the conductive material is not particularly limited. The conductive material particles may have a spherical shape or a scale-like shape, or the 10 conductive material may contain spherical conductive material particles and scale-like conductive material particles. The average particle diameter of the conductive material is not particularly limited but a conductive material having an average particle diameter of about 0.1 μm to about 2 μm is normally used for preparing the electrode paste and the 15 conductive material having an average particle diameter of about 0.2 μm to about 1 μm is preferably used for preparing the electrode paste.

The conductive paste preferably contains the binder in an amount about 2.5 weight parts to about 20 weight parts with respect to 100 weight parts of the conductive material.

20 The content of the solvent is preferably about 40 weight % to about 60 weight % with respect to the weight of the conductive paste.

In order to improve adhesion property, it is preferable for the conductive paste to contain a plasticizing agent. The plasticizing agent contained in the conductive paste is not particularly limited and 25 illustrative examples thereof include phthalate ester, adipic acid, phosphate ester, glycols and the like. The conductive paste contains the plasticizing agent preferably in an amount of about 10 weight % to about 300 weight % with respect to 100 weight parts of the binder, more

preferably in an amount of about 10 weight parts to about 200 weight parts. In the case where the amount of the plasticizing agent added to the conductive paste is too large, the strength of the electrode layer tends to be markedly lower.

5 As occasion demands, the conductive paste may contain additives selected from among various dispersing agents accessory ingredient compounds and the like.

[0044]

In the present invention, preferably, prior to forming an electrode 10 layer or after forming an electrode layer and drying it, a dielectric paste adapted for forming a spacer layer and containing ethyl cellulose having an apparent weight average molecular weight of 110,000 to 190,000 as a binder and at least one kind of solvent selected from the group consisting of isobornyl acetate, dihydroterpinal methyl ether, terpinal methyl ether, 15 α -terpinal acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate is printed on the surface of a ceramic green sheet in a complementary pattern to that of the electrode layer using a screen printing machine, a gravure printing machine or the like, thereby forming a spacer layer.

20 [0045]

In the case where a spacer layer is formed on the surface of a ceramic green sheet in a complementary pattern to that of the electrode layer in this manner, it is possible to prevent a step from being formed 25 between the surface of the electrode layer and the surface of the ceramic green sheet where no electrode layer is formed. Therefore, even in the case of laminating a number of multi-layered units each including a ceramic green sheet and an electrode layer and fabricating a multi-layered electronic component such as a multi-layered ceramic

capacitor, it is possible to effectively prevent the thus fabricated multi-layered electronic component from being deformed and also effectively prevent delamination of layers from occurring.

[0046]

5 Furthermore, as described above, since the solvent selected from the group consisting of isobornyl acetate, dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate hardly dissolves the butyral system resin contained in the ceramic green
10 sheet as a binder, it is possible to reliably prevent the ceramic green sheet from being swollen or partially dissolved so as to generate voids at the interface between the ceramic green sheet and the spacer layer or generate fissures or wrinkles on the surface of the spacer layer.

[0047]

15 Moreover, since the dielectric paste containing ethyl cellulose having an apparent weight average molecular weight of 110,000 to 190,000 as a binder and at least one kind of solvent selected from the group consisting of isobornyl acetate, dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate has a viscosity suitable for printing, a spacer layer can be formed on a ceramic green sheet in a complimentary pattern to that of the electrode layer in a desired manner using a screen printing machine, a gravure printing machine or the like.

20 [0048]

It is preferable for a dielectric paste to contain ethyl cellulose having an apparent weight average molecular weight of 115,000 to 180,000 as a binder.

In this embodiment, the dielectric paste for forming the spacer layer is prepared in the similar manner to the dielectric paste for forming the ceramic green sheet except that different binder and solvent are used.

[0049]

5 Then, the electrode layer or the electrode layer and the spacer layer are dried and a multi-layered unit including the ceramic green sheet and electrode layer or the electrode layer and the spacer layer laminated on the support sheet is fabricated.

[0050]

10 When a multi-layered ceramic capacitor is to be fabricated, the support sheet is peeled off from the ceramic green sheet of the multi-layered unit and the multi-layered unit is diced to predetermined dimensions. Then, a predetermined number of the multi-layered units are laminated on the outer layer of a multi-layered ceramic capacitor and the 15 other outer layer of a multi-layered ceramic capacitor is further laminated on the multi-layered units, thereby fabricating a laminated body. Next, the thus obtained laminated body is press molded and diced to predetermined dimensions, thereby fabricating ceramic green chips.

[0051]

20 The thus fabricated ceramic green chips are placed in a reducing gas atmosphere so that the binder is removed therefrom and the ceramic green chips are baked.

[0052]

25 Necessary external electrodes are then attached to the thus baked ceramic green chip, thereby manufacturing a multi-layered ceramic capacitor.

[0053]

According to this embodiment, since the spacer layer is formed on

the surface of the ceramic green sheet in a complementary pattern to that of the electrode layer, it is possible to prevent a step from being formed between the surface of the electrode layer and the surface of the ceramic green sheet where no electrode layer is formed. Therefore, even in the 5 case of laminating a number of multi-layered units each including a ceramic green sheet and an electrode layer and fabricating a multi-layered electronic component such as a multi-layered ceramic capacitor, it is possible to effectively prevent the thus fabricated multi-layered electronic component from being deformed and also 10 effectively prevent delamination of layers from occurring.

[0054]

Further, according to this embodiment, the spacer layer is formed by printing the dielectric paste containing ethyl cellulose having an apparent weight average molecular weight of 110,000 to 190,000 as a 15 binder and at least one kind of solvent selected from the group consisting of isobornyl acetate, dihydroterpinal methyl ether, terpinal methyl ether, α -terpinal acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate on the ceramic green sheet containing a butyral system resin as a binder in a complementary pattern 20 to that of the electrode layer and the solvent selected from the group consisting of isobornyl acetate, dihydroterpinal methyl ether, terpinal methyl ether, α -terpinal acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate hardly 25 dissolves the butyral system resin contained in a ceramic green sheet as a binder. As a result, even in the case of printing the dielectric paste on a very thin ceramic green sheet, thereby forming a spacer layer, it is possible to reliably prevent the binder contained in the ceramic green sheet from being dissolved by the solvent contained in the dielectric paste

and the ceramic green sheet from being swollen or partially dissolved so as to generate voids at the interface between the ceramic green sheet and the spacer layer or generate fissures or wrinkles on the surface of the spacer layer. Therefore, in the case where a multi-layered ceramic 5 capacitor is fabricated by laminating a number of multi-layered units each including a ceramic green sheet and an electrode layer, it is possible to reliably prevent voids from being generated in the multi-layered ceramic capacitor and it is also possible to reliably prevent the portions of the spacer layer where fissures or wrinkles are generated from dropping 10 off during lamination of a number of the multi-layered units to fabricate the laminated body and mixing into the laminated body as a foreign substance so as to cause internal defects in the multi-layered ceramic capacitor.

[0055]

15 Moreover, according to this embodiment, since the electrode layer is formed by printing the conductive paste containing a binder containing ethyl cellulose having a weight average molecular weight of MW_L and ethyl cellulose having a weight average molecular weight of MW_H at a weight ratio of $X : (1-X)$, where MW_L , MW_H and X are selected so that $X^* 20 MW_L + (1-X)^* MW_H$ falls within a range of 155,000 to 205,000 and at least one solvent selected from the group consisting of isobornyl acetate, dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate on the ceramic green sheet containing a butyral 25 system resin as a binder in a predetermined pattern and the solvent selected from the group consisting of isobornyl acetate, dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl

acetate hardly dissolves the butyral system resin contained in a ceramic green sheet as a binder. As a result, even in the case of printing the conductive paste on a very thin ceramic green sheet, thereby forming an electrode layer, it is possible to reliably prevent the binder contained in 5 the ceramic green sheet from being dissolved by the solvent contained in the conductive paste and the ceramic green sheet from being swollen or partially dissolved. Therefore, even in the case where a ceramic green sheet is very thin, it is possible to effectively prevent generation of pinholes or cracks in the ceramic green sheet and it is therefore possible 10 to effectively prevent short circuit failure from occurring in a multi-layered ceramic capacitor fabricated by laminating a number of multi-layered units.

[0056]

In another preferred embodiment of the present invention, a 15 second support sheet is provided separately from the long support sheet used for forming the ceramic green sheet and the surface of the long second support sheet is coated using a wire bar coater or the like with a dielectric paste containing particles of a dielectric material having substantially the same composition as that of the dielectric material 20 contained in the ceramic green sheet and the same binder as that contained in the ceramic green sheet, thereby forming a coating layer and the coating layer is dried to form a release layer.

[0057]

As the second support sheet, a polyethylene terephthalate film is 25 employed, for example, and the surface of the second support sheet may be coated with a silicon resin, an alkyd resin or the like in order to improve the releasability thereof.

[0058]

The thickness of the release layer is preferably equal to or thinner than that of an electrode layer, more preferably equal to or thinner than about 60 % of the electrode layer thickness and most preferably equal to or thinner than about 30 % of the electrode layer thickness.

5 [0059]

After the release layer has been dried, the conductive paste for an electrode layer prepared in the above described manner is printed on the surface of the release layer in a predetermined pattern using a screen printing machine, a gravure printing machine or the like, thereby forming 10 an electrode layer.

[0060]

It is preferable to form the electrode layer so as to have a thickness of about 0.1 μm to about 5 μm and it is more preferable to form the electrode layer so as to have a thickness of about 0.1 μm to about 1.5 μm .

15 [0061]

In this embodiment, the conductive paste contains a binder containing ethyl cellulose having a weight average molecular weight of MW_L and ethyl cellulose having a weight average molecular weight of MW_H at a weight ratio of $X : (1-X)$, where MW_L , MW_H and X are selected 20 so that $X^* \text{MW}_L + (1-X)^* \text{MW}_H$ falls within a range of 155,000 to 205,000 and at least one solvent selected from the group consisting of isobornyl acetate, dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate.

25 [0062]

Since the solvent selected from the group consisting of isobornyl acetate, dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl

acetate and I-carvyl acetate hardly dissolves a butyral system resin contained in a ceramic green sheet as a binder, even in the case of forming a release layer containing the same binder as that of the ceramic green sheet and printing the conductive paste on the release layer to form an electrode layer, it is possible to effectively prevent the release layer from being swollen or partially dissolved so as to generate voids at the interface between the release layer and the electrode layer or generate fissures or wrinkles on the surface of the electrode layer.

[0063]

Further, since the conductive paste containing a binder containing ethyl cellulose having a weight average molecular weight of MW_L and ethyl cellulose having a weight average molecular weight of MW_H at a weight ratio of $X : (1-X)$, where MW_L , MW_H and X are selected so that $X^* MW_L + (1-X)^* MW_H$ falls within a range of 155,000 to 205,000 and at least one solvent selected from the group consisting of isobornyl acetate, dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate has a viscosity suitable for printing, an electrode layer can be formed on the ceramic green sheet in a predetermined pattern using a screen printing machine, a gravure printing machine or the like in a desired manner.

[0064]

In the present invention, preferably, prior to forming an electrode layer or after forming an electrode layer and drying it, a dielectric paste containing a binder containing ethyl cellulose having an apparent weight average molecular weight of 110,000 to 190,000 as a binder and at least one kind of solvent selected from the group consisting of isobornyl acetate, dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl acetate,

I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate and prepared in the above described manner is printed on the surface of the release layer in a complementary pattern to that of the electrode layer using a screen printing machine, a gravure printing machine or the like, thereby forming a spacer layer.

5 [0065]

In the case where the spacer layer is formed on the surface of a release layer in a complementary pattern to that of the electrode layer in this manner, it is possible to prevent a step from being formed between 10 the surface of the electrode layer and the surface of the release layer where no electrode layer is formed. Therefore, even in the case of laminating a number of multi-layered units each including a ceramic green sheet and an electrode layer and fabricating a multi-layered electronic component such as a multi-layered ceramic capacitor, it is 15 possible to effectively prevent the thus fabricated multi-layered electronic component from being deformed and also effectively prevent delamination of layers from occurring.

15 [0066]

Further, as described above, since the solvent selected from the 20 group consisting of isobornyl acetate, dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate hardly dissolves the butyral system resin contained in the ceramic green 25 sheet as a binder, even in the case of forming the release layer containing the same binder as that of the ceramic green sheet and printing a dielectric paste on the release layer to form a spacer layer, it is possible to effectively prevent the release layer from being swollen or partially dissolved so as to generate voids at the interface between the release

layer and the spacer layer or generate fissures or wrinkles on the surface of the spacer layer.

[0067]

Further, since the dielectric paste containing a binder containing 5 ethyl cellulose having an apparent weight average molecular weight of 110,000 to 190,000 as a binder and at least one kind of solvent selected from the group consisting of isobornyl acetate, dihydroterpinal methyl ether, terpinal methyl ether, α -terpinal acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate has 10 a viscosity suitable for printing, a spacer layer can be formed on the surface of the release layer in a complementary pattern to that of the electrode layer using a screen printing machine, a gravure printing machine or the like in a desired manner.

[0068]

15 Further, a long third support sheet is provided and the surface of the third support sheet is coated with an adhesive agent solution using a bar coater, an extrusion coater, a reverse coater, a dip coater, a kiss coater or the like and the coating layer is dried, thereby forming an adhesive layer.

20 [0069]

It is preferable for the adhesive agent solution to contain a binder belonging to the same group as that the binder contained in the ceramic green sheet belongs to, particles of a dielectric material having substantially the same composition as that of dielectric particles 25 contained in the ceramic green sheet, a plasticizing agent, an antistatic agent and a release agent.

[0070]

It is preferable to form an adhesive layer so as to have a thickness

thinner than about 0.3 μm , more preferable to form it so as to have a thickness of about 0.02 μm to about 0.3 μm and particularly preferable to form it so as to have a thickness of about 0.02 μm to about 0.2 μm .

[0071]

5 The adhesive layer formed on the long third support sheet in this manner is bonded onto the surface of the electrode layer or the surfaces of the electrode layer and the spacer layer formed on the long second support sheet or the surface of the ceramic green sheet formed on the support sheet and the third support sheet then is peeled off from the 10 adhesive layer, whereby the adhesive layer is transferred onto the surface of the electrode layer or the surfaces of the electrode layer and the spacer layer or the surface of the ceramic green sheet.

[0072]

15 In the case where the adhesive layer is transferred onto the surface of the electrode layer or the surfaces of the electrode layer and the spacer layer, the ceramic green sheet formed on the long support sheet is bonded onto the adhesive layer and the first support sheet is peeled off from the ceramic green sheet so that the ceramic green sheet is transferred onto the surface of the adhesive layer, thereby fabricating a 20 multi-layered unit including the ceramic green sheet and the electrode layer or the electrode layer and the spacer layer.

[0073]

25 An adhesive layer is transferred onto the surface of the ceramic green sheet of the thus fabricated multi-layered unit in a similar manner to that of transferring the adhesive layer onto the surface of the electrode layer or the surfaces of the electrode layer and the spacer layer and the multi-layered unit including the adhesive layer transferred onto the surface thereof is diced to predetermined dimensions.

[0074]

Similarly, a predetermined number of multi-layered units each including the adhesive layer transferred onto the surface thereof are fabricated and the predetermined number of multi-layered units are 5 laminated, thereby fabricating a multi-layered block.

[0075]

When a multi-layered block is to be fabricated, the multi-layered unit is first positioned on a support formed of polyethylene terephthalate or the like in such a manner that the adhesive layer transferred onto the 10 surface of the multi-layered unit comes into contact with the support and the multi-layered unit is pressed by a pressing machine or the like, whereby the multi-layered unit is bonded onto the support via the adhesive layer.

[0076]

15 Afterwards, the second support sheet is peeled off from the release layer and the multi-layered unit is laminated on the support.

[0077]

Then, a new multi-layered unit is positioned on the surface of the release layer of the multi-layered unit laminated on the support in such a 20 manner that an adhesive layer formed on the new multi-layered unit comes into contact with the surface of the release layer and the multi-layered unit is pressed using a pressing machine or the like, whereby the new multi-layered unit is laminated on the surface of the release layer of the multi-layered unit laminated on the support via the 25 adhesive layer. Afterwards, the second support sheet is peeled off from the release layer of the new multi-layered unit.

[0078]

Similar processes are repeated, thereby fabricating a

multi-layered block including a predetermined number of the laminated multi-layered units.

[0079]

On the other hand, in the case where the adhesive layer is 5 transferred onto the surface of the ceramic green sheet, the electrode layer or the electrode layer and the spacer layer formed on the second support sheet are bonded onto the adhesive layer and then, the second support sheet is peeled off from the release layer, the electrode layer or the electrode layer and the spacer layer and the release layer are 10 transferred onto the surface of the adhesive layer. Thus, a multi-layered unit including the ceramic green sheet and the electrode layer is fabricated.

[0080]

An adhesive layer is transferred onto the surface of the release 15 layer of the thus obtained multi-layered unit in a similar manner to that of transferring the adhesive layer onto the surface of the ceramic green sheet and the multi-layered unit including the adhesive layer transferred onto the surface thereof is diced to predetermined dimensions.

[0081]

20 Similarly, a predetermined number of multi-layered units each including the adhesive layer transferred onto the surface thereof are fabricated and the predetermined number of multi-layered units are laminated, thereby fabricating a multi-layered block.

[0082]

25 When a multi-layered block is to be fabricated, the multi-layered unit is first positioned on a support formed of polyethylene terephthalate or the like in such a manner that the adhesive layer transferred onto the surface of the multi-layered unit comes into contact with the support and

the multi-layered unit is pressed by a pressing machine or the like, whereby the multi-layered unit is bonded onto the support via the adhesive layer.

[0083]

5 Afterwards, the support sheet is peeled off from the ceramic green sheet and the multi-layered unit is laminated on the support.

[0084]

Then, a new multi-layered unit is positioned on the surface of the ceramic green sheet of the multi-layered unit laminated on the support in 10 such a manner that an adhesive layer formed on the new multi-layered unit comes into contact with the surface of the ceramic green sheet and the multi-layered unit is pressed using a pressing machine or the like, whereby the new multi-layered unit is laminated on the surface of the ceramic green sheet of the multi-layered unit laminated on the support 15 via the adhesive layer. Afterwards, the support sheet is peeled off from the release layer of the new multi-layered unit.

[0085]

Similar processes are repeated, thereby fabricating a multi-layered block including a predetermined number of the laminated 20 multi-layered units.

[0086]

The thus fabricated multi-layered block including the predetermined number of the laminated multi-layered units is laminated on the outer layer of a multi-layered ceramic capacitor and the other outer 25 layer of a multi-layered ceramic capacitor is further laminated on the multi-layered block, thereby fabricating a laminated body. Next, the thus obtained laminated body is press molded and diced to predetermined dimensions, thereby fabricating a number of ceramic green chips.

[0087]

The thus fabricated ceramic green chips are placed in a reducing gas atmosphere so that the binder is removed therefrom and the ceramic green chips are baked.

5 [0088]

Necessary external electrodes are then attached to the thus baked ceramic green chip, thereby manufacturing a multi-layered ceramic capacitor.

[0089]

10 According to this preferred embodiment, since the electrode layer and the spacer layer formed on the second support sheet are dried and then bonded onto the surface of the ceramic green sheet via the adhesive layer, unlike in the case of printing a conductive paste on the surface of the ceramic green sheet to form an electrode layer and printing a dielectric paste on the surface of the ceramic green sheet to form a spacer 15 layer, it is possible to prevent the conductive paste and the dielectric paste from permeating into the ceramic green sheet and it is therefore possible to laminate the electrode layer and the spacer layer on the surface of the ceramic green sheet in a desired manner.

20 [0090]

Further, according to this preferred embodiment, the spacer layer is formed using the dielectric paste containing a binder containing ethyl cellulose having an apparent weight average molecular weight of 110,000 to 190,000 as a binder and at least one kind of solvent selected from the 25 group consisting of isobornyl acetate, dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate and the solvent selected from the group consisting of isobornyl acetate,

dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate hardly dissolves a butyral system resin contained in a ceramic green sheet as a binder. As a result, even in the case of 5 forming a release layer containing the same binder as that contained in the ceramic green sheet and printing a dielectric paste on the surface of the release layer, thereby forming a spacer layer, it is possible to effectively prevent the release layer from being swollen or partially dissolved so as to generate voids at the interface between the release 10 layer and the spacer layer or generate fissures or wrinkles on the surface of the spacer layer. Therefore, in the case where a multi-layered ceramic capacitor is fabricated by laminating a number of multi-layered units each including a ceramic green sheet and an electrode layer, it is possible to reliably prevent voids from being generated in the multi-layered 15 ceramic capacitor and it is also possible to reliably prevent the portions of the spacer layer where fissures or wrinkles are generated from dropping off during lamination of a number of the multi-layered units to fabricate the laminated body and mixing into the laminated body as a foreign substance so as to cause internal defects in the multi-layered ceramic 20 capacitor.

[0091]

Furthermore, according to this preferred embodiment, the electrode layer is formed using the conductive paste containing a binder containing ethyl cellulose having a weight average molecular weight of 25 MW_L and ethyl cellulose having a weight average molecular weight of MW_H at a weight ratio of $X : (1-X)$, where MW_L , MW_H and X are selected so that $X * MW_L + (1-X) * MW_H$ falls within a range of 155,000 to 205,000 and at least one solvent selected from the group consisting of isobornyl

acetate, dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl acetate, β -dihydrocarvyl acetate, β -menthyl acetate, β -menthone, β -perillyl acetate and β -carvyl acetate and the solvent selected from the group consisting of isobornyl acetate, dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl acetate, β -dihydrocarvyl acetate, β -menthyl acetate, β -menthone, β -perillyl acetate and β -carvyl acetate hardly dissolves a butyral system resin contained in a ceramic green sheet as a binder. As a result, even in the case of forming the release layer containing the same binder as that contained in a ceramic green sheet and printing the conductive paste on the release layer, thereby forming an electrode layer, it is possible to effectively prevent the release layer from being swollen or partially dissolved so as to generate pinholes or cracks in the release layer and effectively prevent defects from being generated in a multi-layered ceramic capacitor.

15 [0092]

Moreover, according to this preferred embodiment, since it is possible to prevent the release layer from being swollen or partially dissolved, thereby changing the release strength between the release layer and the electrode layer and the spacer layer or the release layer and the electrode layer, it is possible to effectively prevent defects from being generated when a multi-layered unit is fabricated.

[0093]

In a further preferred embodiment, in the case where the adhesive layer is transferred onto the surface of the electrode layer or the surfaces 25 of the electrode layer and the spacer layer, an adhesive layer is transferred onto the surface of a ceramic green sheet of a multi-layered unit fabricated by laminating a release layer, an electrode layer or an electrode layer and a spacer layer, an adhesive layer and a ceramic green

sheet on a long second support sheet and without cutting the multi-layered unit, a release layer of another multi-layered unit fabricated by laminating a ceramic green sheet, an adhesive layer, an electrode layer or an electrode layer and a spacer layer, and the release 5 layer on a long support sheet is bonded onto the adhesive layer and the support sheet is peeled off from the ceramic green sheet, whereby two multi-layered units are laminated on the long second support sheet.

[0094]

Then, an adhesive layer formed on a third support sheet is 10 transferred onto the ceramic green sheet located on the side of the surface of the laminated two multi-layered units and a release layer of another multi-layered unit fabricated by laminating a ceramic green sheet, an adhesive layer, an electrode layer or an electrode layer and a spacer layer, and the release layer on a long support sheet is bonded onto the adhesive 15 layer and the support sheet is peeled off from the release layer.

[0095]

Similar processes are repeated, thereby fabricating a multi-layered unit set including a predetermined number of laminated multi-layered units. Further, an adhesive layer formed on the third support sheet is transferred onto the surface of the ceramic green sheet located on the side of the surface of the multi-layered unit set, thereby 20 fabricating a laminated body and the laminated body is diced to predetermined dimensions, thereby fabricating a multi-layered blocks.

[0096]

On the other hand, in the case where the adhesive layer is 25 transferred onto the surface of the ceramic green sheet, an adhesive layer is transferred onto the surface of a release layer of a multi-layered unit fabricated by laminating a ceramic green sheet, an adhesive layer, an

electrode layer or an electrode layer and a spacer layer, and the release layer on a long support sheet and without cutting the multi-layered unit, a ceramic green sheet of another multi-layered unit fabricated by laminating a release layer, an electrode layer or an electrode layer and a 5 spacer layer, an adhesive layer and a ceramic green sheet on a long second support sheet is bonded onto the adhesive layer and the second support sheet is peeled off from the release layer, whereby two multi-layered units are laminated on the long second support sheet.

[0097]

10 Then, an adhesive layer formed on a third support sheet is transferred onto the release layer located on the side of the surface of the laminated two multi-layered units and a ceramic green sheet of a multi-layered unit fabricated by laminating a release layer, an electrode layer or an electrode layer and a spacer layer, an adhesive layer and a 15 ceramic green sheet on a long second support sheet is further laminated on the adhesive layer. Then, the second support sheet is peeled off from the release layer.

[0098]

Similar processes are repeated, thereby fabricating a 20 multi-layered unit set including a predetermined number of laminated multi-layered units. Further, an adhesive layer formed on the third support sheet is transferred onto the surface of the release layer located on the side of the surface of the multi-layered unit set, thereby fabricating a laminated body and the laminated body is diced to predetermined 25 dimensions, thereby fabricating multi-layered blocks.

[0099]

A multi-layered ceramic capacitor is fabricated using the thus fabricated multi-layered blocks in the manner of the previous preferred

embodiment.

[0100]

According to this preferred embodiment, since the multi-layered units are successively laminated on the long second support sheet or support sheet, thereby fabricating the multi-layered unit set including a predetermined number of multi-layered units and the multi-layered unit set is diced to predetermined dimensions, thereby fabricating multi-layered blocks, it is possible to markedly improve the manufacturing efficiency of the multi-layered blocks in comparison with the case where multi-layered blocks are fabricated by laminating multi-layered units each of which has been diced to predetermined dimensions.

[0101]

In a further preferred embodiment of the present invention, in the case where the adhesive layer is transferred onto the surface of the electrode layer or the surfaces of the electrode layer and the spacer layer, an adhesive layer is transferred onto the surface of a ceramic green sheet of a multi-layered unit fabricated by laminating a release layer, an electrode layer or an electrode layer and a spacer layer, an adhesive layer and a ceramic green sheet on a long second support sheet and without cutting the multi-layered unit, an electrode layer or an electrode layer and a spacer layer formed on the second support sheet are bonded onto the adhesive layer and the second support sheet is peeled off from the release layer, whereby the electrode layer and the spacer layer, and the release layer are transferred onto the surface of the adhesive layer.

[0102]

Then, an adhesive layer formed on a third support sheet is transferred onto the surface of the release layer transferred onto the

adhesive layer, a ceramic green sheet formed on the support sheet is bonded onto the adhesive layer and the support sheet is peeled off from the ceramic green sheet, whereby the ceramic green sheet is transferred onto the surface of the adhesive layer.

5 [0103]

Further, an adhesive layer formed on a third support sheet is transferred onto the surface of the ceramic green sheet transferred onto the surface of the adhesive layer, an electrode layer or an electrode layer and a spacer layer formed on the second support sheet are bonded onto the adhesive layer and the second support sheet is peeled off from the release layer, whereby the electrode layer or the electrode layer and the spacer layer, and the release layer are transferred onto the surface of the adhesive layer.

10 [0104]

15 Similar processes are repeated, thereby fabricating a multi-layered unit set including a predetermined number of laminated multi-layered units. Further, an adhesive layer formed on the third support sheet is transferred onto the surface of the ceramic green sheet located on the side of the surface of the multi-layered unit set, thereby 20 fabricating a laminated body and the laminated body is diced to predetermined dimensions, thereby fabricating multi-layered blocks.

[0105]

On the other hand, in the case where the adhesive layer is transferred onto the surface of the ceramic green sheet, an adhesive layer 25 is transferred onto the surface of a release layer of a multi-layered unit fabricated by laminating a ceramic green sheet, an adhesive layer, an electrode layer or an electrode layer and a spacer layer, and the release layer on a long support sheet and without cutting the multi-layered unit,

a ceramic green sheet of a support sheet is bonded onto the adhesive layer and the support sheet is peeled off from the ceramic green sheet, whereby the ceramic green sheet is transferred onto the adhesive layer.

[0106]

5 Further, an adhesive layer formed on the third support sheet is transferred onto the ceramic green sheet transferred onto the adhesive layer and an electrode layer or an electrode layer and a spacer layer formed on the second support sheet are bonded onto the adhesive layer. Then, the second support sheet is peeled off from the release layer, 10 whereby the electrode layer or the electrode layer and the spacer layer, and the release layer are transferred onto the surface of the adhesive layer.

[0107]

15 Further, an adhesive layer formed on the third support sheet is transferred onto the release layer transferred onto the adhesive layer and a ceramic green sheet formed on the support sheet is bonded onto the adhesive layer. Then, the support sheet is peeled off from the ceramic green sheet, whereby the ceramic green sheet is transferred onto the surface of the adhesive layer.

20 [0108]

Similar processes are repeated, thereby fabricating a multi-layered unit set including a predetermined number of laminated multi-layered units. Further, an adhesive layer is transferred onto the surface of the release layer located on the side of the surface of the multi-layered unit set, thereby fabricating a laminated body and the laminated body is diced to predetermined dimensions, thereby fabricating multi-layered blocks.

[0109]

A multi-layered ceramic green sheet is fabricated using the thus fabricated multi-layered block in the manner of the previous embodiment.

[0110]

According to this preferred embodiment, the transferring of the adhesive layer, the transferring of the electrode layer or the electrode layer and the spacer layer and the release layer, the transferring of the adhesive layer and the transferring of the ceramic green sheet onto the long second support sheet or support sheet are repeated, thereby successively laminating the multi-layered units to fabricate the multi-layered unit set including a predetermined number of multi-layered units and the multi-layered unit set is diced to predetermined dimensions, thereby fabricating multi-layered blocks. As a result, it is possible to markedly improve the manufacturing efficiency of the multi-layered block in comparison with the case where multi-layered blocks are fabricated by laminating multi-layered units each of which has been diced to predetermined dimensions.

[WORKING EXAMPLES]

[0111]

Hereinafter, working examples and comparative examples will be set out in order to further clarify the advantages of the present invention.

[0112]

Working Example 1

Preparation of a dielectric paste for forming a ceramic green sheet

1.48 weight parts of $(\text{BaCa})\text{SiO}_3$, 1.01 weight parts of Y_2O_3 , 0.72 weight part of MgCO_3 , 0.13 weight part of MnO and 0.045 weight part of V_2O_5 were mixed, thereby preparing an additive powder.

[0113]

72.3 weight parts of ethyl alcohol, 72.3 weight parts of propyl alcohol, 25.8 weight parts of xylene and 0.93 weight parts of polyethyleneglycol system dispersing agent were added to 100 weight parts of the thus prepared additive powder to prepare a slurry and the 5 additives contained in the slurry were pulverized.

[0114]

When the additives contained in the slurry were to be pulverized, 11.65 grams of the slurry and 450 grams of ZrO_2 beads having a diameter of 2 mm were charged in a polyethylene vessel having an inner volume of 10 250 cc and the polyethylene vessel was rotated at the circumferential velocity of 45 m/min for sixteen hours, thereby pulverizing the additive powder to prepare the additive slurry.

[0115]

The median diameter of the additives after pulverization was 0.1 15 μm .

[0116]

Then, 15 weight parts of polyvinyl butyral whose degree of polymerization was 1450 and degree of butyralization was 69 mol % was dissolved into a mixture of 42.5 weight parts of ethyl alcohol and 42.5 20 weight parts of propyl alcohol at 50 °C, thereby preparing a 15 % organic vehicle solution. Further, a slurry having the composition set out below was mixed with the organic vehicle solution for twenty hours using a ball mill having an inner volume of 500 cc, thereby preparing a dielectric paste. When the slurry was to be mixed with the organic vehicle solution, 25 330.1 grams of the slurry and 900 grams of ZrO_2 beads having a diameter of 2 mm were charged in the polyethylene vessel and the polyethylene vessel was rotated at the circumferential velocity of 45 m/min.

[0117]

BaTiO₃ powders ("BT-02" (Product Name) manufactured by SAKAI CHEMICAL INDUSTRY CO., LTD.: particle diameter 0.2 µm)

		100 weight parts
	additive slurry	11.65 weight parts
5	ethyl alcohol	35.32 weight parts
	propyl alcohol	35.32 weight parts
	xylene	16.32 weight parts
	benzyl butyl phthalate (plasticizing agent)	2.61 weight parts
	mineral spirit	7.3 weight parts
10	polyethylene glycol system dispersing agent	2.36 weight parts
	imidazoline system antistatic auxiliary agent	0.42 weight parts
	organic vehicle	33.74 weight parts
	methyl ethyl ketone	43.81 weight parts
	2-butoxyethyl alcohol	43.81 weight parts

15

As a polyethylene glycol system dispersing agent, a dispersing agent which was obtained by denaturing polyethylene glycol with aliphatic acid and whose hydrophile-liophile balance (HLB) was 5 to 6 was employed.

20

[0118]

Formation of a ceramic green sheet

A polyethylene terephthalate film was coated with the thus prepared dielectric paste using a die coater at a coating velocity of 50 m/minutes, thereby forming a coating layer and the thus formed coating layer was dried in a drying furnace whose temperature was held at 80 °C, thereby forming a ceramic green sheet having a thickness of 1 µm.

25

[0119]

Formation of a ceramic green sheet

A polyethylene terephthalate film was coated with the thus prepared dielectric paste using a die coater at a coating velocity of 50 m/minutes, thereby forming a coating layer and the thus formed coating layer was dried in a drying furnace whose temperature was held at 80 °C, 5 thereby forming a ceramic green sheet having a thickness of 1 μm .

[0120]

Preparation of a dielectric paste for forming an electrode layer

1.48 weight parts of $(\text{BaCa})\text{SiO}_3$, 1.01 weight parts of Y_2O_3 , 0.72 weight part of MgCO_3 , 0.13 weight part of MnO and 0.045 weight part of 10 V_2O_5 were mixed, thereby preparing an additive powder.

[0121]

150 weight parts of acetone, 104.3 weight parts of isobornyl acetate and 1.5 weight parts of polyethylene glycol system dispersing agent were added to 100 weight parts of the thus prepared additive 15 powder to prepare a slurry and the additives contained in the slurry were pulverized using a pulverizer "LMZ0.6" (Product name) manufactured by Ashizawa Finetech Co., Ltd.

[0122]

When the additives contained in the slurry were to be pulverized, 20 ZrO_2 beads having a diameter of 0.1 mm were charged into a vessel so as to occupy 80 volume % of the vessel, a rotor was rotated at the circumferential velocity of 14 m/min and the slurry was circulated between the vessel and a slurry tank until the holding time of the whole 25 slurry became 5 minutes, thereby pulverizing the additives contained in the slurry.

[0123]

The median diameter of the additives after pulverization was 0.1 μm .

[0124]

Then, acetone was evaporated using an evaporator and removed from the slurry, thereby preparing an additive paste in which the additives were dispersed in isobornyl acetate. The concentration of the 5 additives contained in the additive paste was 49.3 weight %.

[0125]

Then, 8 weight parts of a binder containing ethyl cellulose having a weight average molecular weight of 75,000 and ethyl cellulose having a weight average molecular weight of 130,000 at a weight ratio of 25:75, 10 namely, 8 weight parts of ethyl cellulose having an apparent weight average molecular weight of 116,250, was dissolved in 92 weight parts of isobornyl acetate at 70 °C, thereby preparing an 8 % organic vehicle solution. Further, a slurry having the composition set out below was dispersed in the organic vehicle solution for sixteen hours using a ball 15 mill. The dispersing conditions were set so that the amount of charged ZrO_2 having a diameter of 2.0 mm was 30 volume % of the ball mill, the amount of the slurry in the ball mill was 60 volume % and the circumferential velocity of the ball mill was 45 m/min.

[0126]

20	additive paste	8.87 weight parts
	BaTiO ₃ powder (manufactured by SAKAI CHEMICAL INDUSTRY CO., LTD.: particle diameter 0.05 μm)	95.70 weight parts
	organic vehicle	104.36 weight parts
25	polyethylene glycol system dispersing agent	1.00 weight parts
	dioctyl phthalate (plasticizing agent)	2.61 weight parts
	imidazoline system surfactant	0.4 weight parts
	acetone	57.20 weight parts

[0127]

Then, acetone was evaporated using a stirring device having an evaporator and a heating mechanism and removed from the slurry, thereby preparing a dielectric paste.

5 [0128]

The viscosity of the thus obtained dielectric paste was measured using a rheometer manufactured by HAAKE Co., Ltd. under conditions of a temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also measured under conditions of a temperature of 25 °C and shearing

10 velocity of 50 sec⁻¹.

[0129]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec⁻¹ was 7.99 Ps·s and that the viscosity of the dielectric paste measured under condition of 15 the shearing velocity of 50 sec⁻¹ was 4.24 Ps·s.

[0130]

Preparation of a dielectric paste for forming an electrode layer

1.48 weight parts of (BaCa)SiO₃, 1.01 weight parts of Y₂O₃, 0.72 weight part of MgCO₃, 0.13 weight part of MnO and 0.045 weight part of 20 V₂O₅ were mixed, thereby preparing an additive powder.

[0131]

150 weight parts of acetone, 104.3 weight parts of isobornyl acetate and 1.5 weight parts of polyethylene glycol system dispersing agent were added to 100 weight parts of the thus prepared additive 25 powder to prepare a slurry and the additives contained in the slurry were pulverized using a pulverizer "LMZ0.6" (Product name) manufactured by Ashizawa Finetech Co., Ltd.

[0132]

When the additives contained in the slurry were to be pulverized, ZrO₂ beads having a diameter of 0.1 mm were charged into a vessel so as to occupy 80 volume % of the vessel, a rotor was rotated at the circumferential velocity of 14 m/min and the slurry was circulated 5 between the vessel and a slurry tank until the holding time of the whole slurry became 5 minutes, thereby pulverizing the additives contained in the slurry.

[0133]

The median diameter of the additives after pulverization was 0.1 10 μm .

[0134]

Then, acetone was evaporated using an evaporator and removed from the slurry, thereby preparing an additive paste in which the additives were dispersed in terpineol. The concentration of the additives 15 contained in the additive paste was 49.3 weight %.

[0135]

Then, 8 weight parts of a binder containing ethyl cellulose having a weight average molecular weight (MW_L) of 130,000 and ethyl cellulose having a weight average molecular weight (MW_H) of 230,000 at a weight 20 ratio of 50:50, namely, 8 weight parts of ethyl cellulose having an apparent weight average molecular weight of 180,000 defined as $X^*\text{MW}_L + (1-X)^*\text{MW}_H$, was dissolved in 92 weight parts of isobornyl acetate at 70 °C, thereby preparing an 8 % organic vehicle solution. Further, a slurry having the composition set out below was dispersed in the organic vehicle 25 solution for sixteen hours using a ball mill. The dispersing conditions were set so that the amount of charged ZrO₂ having a diameter of 2.0 mm was 30 volume % of the ball mill, the amount of the slurry in the ball mill was 60 volume % and the circumferential velocity of the ball mill was 45

m/min.

[0136]

	nickel powder manufactured by Kawatetsu Industry Co., Ltd. and having a particle diameter of 0.2 μm	100 weight parts
5	additive paste	1.77 weight parts
	BaTiO ₃ powder manufactured by SAKAI CHEMICAL INDUSTRY CO., LTD.	19.14 weight parts
	organic vehicle	56.25 weight parts
	polyethylene glycol system dispersing agent	1.19 weight parts
10	isobornyl acetate	32.19 weight parts
	acetone	56 weight parts

[0137]

Then, acetone was evaporated using a stirring device having an evaporator and a heating mechanism and removed from the slurry, thereby preparing a conductive paste. The concentration of the dielectric material contained in the conductive paste was 47 weight %.

[0138]

Formation of a spacer layer

The thus prepared dielectric paste was printed on the surface of the ceramic green sheet in a predetermined pattern using a screen printing machine and dried at 90 °C for five minutes, thereby forming a spacer layer on the surface of the ceramic green sheet.

[0139]

Further, the surface of the spacer layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the spacer layer was free of cracks and wrinkles.

[0140]

Formation of an electrode layer and fabrication of a multi-layered unit

The thus prepared conductive paste was printed on the ceramic green sheet in a complimentary pattern to that of the spacer layer using a screen printing machine and dried at 90 °C for five minutes, thereby 5 forming an electrode layer having a thickness of 1 μm . Thus, a multi-layered unit including the ceramic green sheet, the electrode layer and the spacer layer laminated on the polyethylene terephthalate film was fabricated.

[0141]

10 Further, the surface of the electrode layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the electrode layer was free of cracks and wrinkles.

[0142]

15 Fabrication of a ceramic green chip

The surface of a polyethylene terephthalate film was coated with the dielectric paste prepared in the above described manner using a die coater, thereby forming a coating layer, and the coating layer was dried, thereby forming a ceramic green sheet having a thickness of 10 μm .

20 [0143]

The thus formed ceramic green sheet was peeled off from the polyethylene terephthalate film and diced. Five of the diced ceramic green sheet units were laminated to form a cover layer having a thickness of 50 μm . Further, the multi-layered unit was peeled off from the polyethylene terephthalate film and diced and fifty of the diced multi-layered units 25 were laminated on the cover layer.

[0144]

Then, the ceramic green sheet having a thickness of 10 μm was

peeled off from the polyethylene terephthalate film and diced and five of the ceramic green sheet units were laminated on the multi-layered units laminated on the cover layer, thereby fabricating a laminated body including the lower cover layer having a thickness of 50 μm , an active 5 layer having a thickness of 100 μm and including the laminated fifty multi-layered units each including the ceramic green sheet having a thickness of 1 μm , the electrode layer having a thickness of 1 μm and the spacer layer having a thickness of 1 μm , and an upper cover layer having a thickness of 50 μm .

10 [0145]

Further, a pressure of 100 MPa was applied onto the thus fabricated laminated body at 70 $^{\circ}\text{C}$, thereby press molding the laminated body and the laminated body was diced to predetermined dimensions using a dicing machine, thereby fabricating ceramic green chips.

15 [0146]

A total of thirty ceramic green chips were fabricated in a manner similar to the foregoing.

[0147]

Baking of ceramic green chips and annealing treatment thereof

20 Each of the thus fabricated ceramic green chip was processed under the following conditions in an air atmosphere to remove the binder.

[0148]

Rate of temperature increase: 50 $^{\circ}\text{C}$ / hour

Holding temperature: 240 $^{\circ}\text{C}$

25 Holding time period: 8 hours

[0149]

After removing the binder, the ceramic green chip was processed and baked under the following conditions in a mixed gas atmosphere of a

nitrogen gas and a hydrogen gas whose temperature was controlled at the dew point 20 °C. The contents of the nitrogen gas and the hydrogen gas contained in the mixed gas were 95 volume % and 5 volume %, respectively.

5 [0150]

Rate of temperature increase: 300 °C / hour

Holding temperature: 1200 °C

Holding time period: 2 hours

Cooling rate: 300 °C / hour

10 [0151]

The thus baked ceramic green chip was subjected to an annealing treatment under the following conditions in a nitrogen gas atmosphere whose temperature was controlled at the dew point 20 °C.

[0152]

15 Rate of temperature increase: 300 °C / hour

Holding temperature: 1000 °C

Holding time period: 3 hours

Cooling rate: 300 °C / hour

[0153]

20 Observation of voids

The ceramic green chip which had been subjected to an annealing treatment in this manner was embedded in a two liquid curable type epoxy resin so that the side surface was exposed to the outside and after the two liquid curable type epoxy resin was cured, the ceramic green chip having a size of 3.2 mm x 1.6 mm was ground by 1.6 mm using # 400 sand paper, # 800 sand paper, # 1000 sand paper and # 2000 sand paper in this order.

[0154]

The thus ground surface of the ceramic green chip was subjected to a mirror polishing processing using 1 μm diamond paste and the thus polished surface of the ceramic green chip was observed at four-hundred magnifications using an optical microscope to examine whether any void 5 was present.

[0155]

As a result, no void was observed in any of thirty ceramic green chips.

[0156]

10 Working Example 2

A dielectric paste for forming a spacer layer was prepared in the manner of Working Example 1 except that ethyl cellulose having a weight average molecular weight of 130,000 was used as a binder of the dielectric paste and the viscosity of the thus prepared conductive paste was 15 measured under conditions of a temperature of 25 $^{\circ}\text{C}$ and shearing velocity of 8 sec^{-1} and was also measured under conditions of a temperature of 25 $^{\circ}\text{C}$ and shearing velocity of 50 sec^{-1} .

[0157]

As a result, it was found that the viscosity of the dielectric paste 20 measured under condition of the shearing velocity of 8 sec^{-1} was 12.8 $\text{Ps}\cdot\text{s}$ and that the viscosity of the dielectric paste measured under condition of the shearing velocity of 50 sec^{-1} was 6.45 $\text{Ps}\cdot\text{s}$.

[0158]

Then, the thus prepared dielectric paste was printed using a 25 screen printing machine on a ceramic green sheet formed in the manner of Working Example 1, thereby forming a spacer layer.

[0159]

Further, the surface of the thus formed spacer layer was observed

at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the spacer layer was free of cracks and wrinkles.

[0160]

5 A conductive paste for forming an electrode layer was then prepared in the manner of Working Example 1 and the thus prepared conductive paste was printed on the ceramic green sheet, thereby fabricating a multi-layered unit including the electrode layer and the spacer layer laminated on the ceramic green sheet.

10 [0161]

Further, the surface of the thus formed electrode layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the electrode layer was free of cracks and wrinkles.

15 [0162]

Furthermore, a total of thirty annealing treated ceramic green chips were fabricated in the manner of Working Example 1 and the surface of each of the ceramic green chips was observed in the manner of Working Example 1. As a result, no void was observed in any of the 20 ceramic green chips.

[0163]

Working Example 3

A dielectric paste for forming a spacer layer was prepared in the manner of Working Example 1 except that ethyl cellulose having a weight 25 average molecular weight of 130,000 and ethyl cellulose having a weight average molecular weight of 230,000 at a weight ratio of 75:25, namely, ethyl cellulose having an apparent weight average molecular weight of 155,000, was used as a binder of the dielectric paste and the viscosity of

the thus prepared conductive paste was measured under conditions of a temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also measured under conditions of a temperature of 25 °C and shearing velocity of 50 sec⁻¹.

5 [0164]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec⁻¹ was 15.1 Ps·s and that the viscosity of the dielectric paste measured under condition of the shearing velocity of 50 sec⁻¹ was 7.98 Ps·s.

10 [0165]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner of Working Example 1, thereby forming a spacer layer.

[0166]

15 Further, the surface of the thus formed spacer layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the spacer layer was free of cracks and wrinkles.

[0167]

20 A conductive paste for forming an electrode layer was then prepared in the manner of Working Example 1 and the thus prepared conductive paste was printed on the ceramic green sheet, thereby fabricating a multi-layered unit including the electrode layer and the spacer layer laminated on the ceramic green sheet.

25 [0168]

Further, the surface of the thus formed electrode layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the electrode

layer was free of cracks and wrinkles.

[0169]

Furthermore, a total of thirty annealing treated ceramic green chips were fabricated in the manner of Working Example 1 and the 5 surface of each of the ceramic green chips was observed in the manner of Working Example 1. As a result, no void was observed in any of the ceramic green chips.

[0170]

Working Example 4

10 A dielectric paste for forming a spacer layer was prepared in the manner of Working Example 1 except that ethyl cellulose having a weight average molecular weight of 130,000 and ethyl cellulose having a weight average molecular weight of 230,000 at a weight ratio of 50:50, namely, ethyl cellulose having an apparent weight average molecular weight of 15 180,000, was used as a binder of the dielectric paste and the viscosity of the thus prepared conductive paste was measured under conditions of a temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also measured under conditions of a temperature of 25 °C and shearing velocity of 50 sec⁻¹.

20 [0171]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec⁻¹ was 19.9 Ps·s and that the viscosity of the dielectric paste measured under condition of the shearing velocity of 50 sec⁻¹ was 10.6 Ps·s.

25 [0172]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner of Working Example 1, thereby forming a spacer layer.

[0173]

Further, the surface of the thus formed spacer layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the spacer layer was free of cracks 5 and wrinkles.

[0174]

A conductive paste for forming an electrode layer was then prepared in the manner of Working Example 1 and the thus prepared conductive paste was printed on the ceramic green sheet, thereby 10 fabricating a multi-layered unit including the electrode layer and the spacer layer laminated on the ceramic green sheet.

[0175]

Further, the surface of the thus formed electrode layer was observed at four-hundred magnifications using a metallographic 15 microscope. As a result, it was found that the surface of the electrode layer was free of cracks and wrinkles.

[0176]

Furthermore, a total of thirty annealing treated ceramic green chips were fabricated in the manner of Working Example 1 and the 20 surface of each of the ceramic green chips was observed in the manner of Working Example 1. As a result, no void was observed in any of the ceramic green chips.

[0177]

Comparative Example 1

25 A dielectric paste for forming a spacer layer was prepared in the manner of Working Example 1 except that ethyl cellulose having a weight average molecular weight of 75,000 and ethyl cellulose having a weight average molecular weight of 130,000 at a weight ratio of 50:50, namely,

ethyl cellulose having an apparent weight average molecular weight of 102,500, was used as a binder of the dielectric paste and the viscosity of the thus prepared conductive paste was measured under conditions of a temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also 5 measured under conditions of a temperature of 25 °C and shearing velocity of 50 sec⁻¹.

[0178]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec⁻¹ was 4.61 Ps·s 10 and that the viscosity of the dielectric paste measured under condition of the shearing velocity of 50 sec⁻¹ was 2.89 Ps·s.

[0179]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner 15 of Working Example 1. However, since the viscosity of the dielectric paste was too low, a spacer layer could not be formed.

[0180]

Comparative Example 2

A dielectric paste for forming a spacer layer was prepared in the 20 manner of Working Example 1 except that ethyl cellulose having a weight average molecular weight of 130,000 and ethyl cellulose having a weight average molecular weight of 230,000 at a weight ratio of 25:75, namely, ethyl cellulose having an apparent weight average molecular weight of 205,000, was used as a binder of the dielectric paste and the viscosity of 25 the thus prepared conductive paste was measured under conditions of a temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also measured under conditions of a temperature of 25 °C and shearing velocity of 50 sec⁻¹.

[0181]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec^{-1} was $25.4 \text{ Ps}\cdot\text{s}$ and that the viscosity of the dielectric paste measured under condition of 5 the shearing velocity of 50 sec^{-1} was $14.6 \text{ Ps}\cdot\text{s}$.

[0182]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner of Working Example 1. However, since the viscosity of the dielectric paste 10 was too high, the clogging of a screen printing plate occurred and a continuous spacer layer could not be formed.

[0183]

Comparative Example 3

A dielectric paste for forming a spacer layer was prepared in the 15 manner of Working Example 1 except that ethyl cellulose having a weight average molecular weight of 230,000 was used as a binder of the dielectric paste and the viscosity of the thus prepared conductive paste was measured under conditions of a temperature of 25°C and shearing velocity of 8 sec^{-1} and was also measured under conditions of a 20 temperature of 25°C and shearing velocity of 50 sec^{-1} .

[0184]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec^{-1} was $34.4 \text{ Ps}\cdot\text{s}$ and that the viscosity of the dielectric paste measured under condition of 25 the shearing velocity of 50 sec^{-1} was $19.2 \text{ Ps}\cdot\text{s}$.

[0185]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner

of Working Example 1. However, since the viscosity of the dielectric paste was too high, the clogging of a screen printing plate occurred and a continuous spacer layer could not be formed.

[0186]

5 Comparative Example 4

A dielectric paste for forming a ceramic green sheet was prepared in the manner of Working Example 1 except that butyral system resin whose degree of polymerization was 800 and degree of butyralization was 69 mol % was used as a binder of the dielectric paste for forming a 10 ceramic green sheet, thereby forming a ceramic green sheet.

Further, the thus prepared dielectric paste was printed using a screen printing machine in the manner of Working Example 4 on a ceramic green sheet formed in the manner of Working Example 1, thereby forming a spacer layer.

15 Then, the surface of the thus formed spacer layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that cracks and wrinkles were generated on the surface of the spacer layer.

[0187]

20 A conductive paste for forming an electrode layer was then prepared in the manner of Working Example 1 and the thus prepared conductive paste was printed on the ceramic green sheet, thereby fabricating a multi-layered unit including the electrode layer and the spacer layer laminated on the ceramic green sheet.

25 [0188]

Further, the surface of the thus formed electrode layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that cracks and wrinkles were

generated on the surface of the electrode layer.

[0189]

Furthermore, a total of thirty annealing treated ceramic green chips were fabricated in the manner of Working Example 1 and the 5 surface of each of the ceramic green chips was observed in the manner of Working Example 1. As a result, voids were observed in the two ceramic green chips among the thirty ceramic green chips.

[0190]

Working Example 5

10 A dielectric paste was prepared in the manner of Working Example 1 except that dihydroterpinyl methyl ether was used as a solvent instead of isobornyl acetate and the viscosity of the thus prepared dielectric paste was measured under conditions of a temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also measured under conditions of a 15 temperature of 25 °C and shearing velocity of 50 sec⁻¹.

[0191]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec⁻¹ was 7.76 Ps·s and that the viscosity of the dielectric paste measured under condition of 20 the shearing velocity of 50 sec⁻¹ was 4.39 Ps·s.

[0192]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner of Working Example 1, thereby forming a spacer layer.

25 [0193]

Further, the surface of the thus formed spacer layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the spacer layer was free of cracks

and wrinkles.

[0194]

A conductive paste for forming an electrode layer was then prepared in the manner of Working Example 1 except that 5 dihydroterpinyl methyl ether was used as a solvent instead of isobornyl acetate and the thus prepared conductive paste was printed on the ceramic green sheet, thereby fabricating a multi-layered unit including the electrode layer and the spacer layer laminated on the ceramic green sheet.

10 [0195]

Further, the surface of the thus formed electrode layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the electrode layer was free of cracks and wrinkles.

15 [0196]

Furthermore, a total of thirty annealing treated ceramic green chips were fabricated in the manner of Working Example 1 and the surface of each of the ceramic green chips was observed in the manner of Working Example 1. As a result, no void was observed in any of the 20 ceramic green chips.

[0197]

Working Example 6

A dielectric paste for forming a spacer layer was prepared in the manner of Working Example 5 except that ethyl cellulose having a weight 25 average molecular weight of 130,000 was used as a binder of the dielectric paste and the viscosity of the thus prepared conductive paste was measured under conditions of a temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also measured under conditions of a

temperature of 25 °C and shearing velocity of 50 sec⁻¹.

[0198]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec⁻¹ was 11.4 Ps·s and that the viscosity of the dielectric paste measured under condition of the shearing velocity of 50 sec⁻¹ was 6.05 Ps·s.

[0199]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner 10 of Working Example 1, thereby forming a spacer layer.

[0200]

Further, the surface of the thus formed spacer layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the spacer layer was free of cracks 15 and wrinkles.

[0201]

A conductive paste for forming an electrode layer was then prepared in the manner of Working Example 1 except that dihydroterpinyl methyl ether was used as a solvent instead of isobornyl acetate and the thus prepared conductive paste was printed on the 20 ceramic green sheet, thereby fabricating a multi-layered unit including the electrode layer and the spacer layer laminated on the ceramic green sheet.

[0202]

Further, the surface of the thus formed electrode layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the electrode 25 layer was free of cracks and wrinkles.

[0203]

Furthermore, a total of thirty annealing treated ceramic green chips were fabricated in the manner of Working Example 1 and the surface of each of the ceramic green chips was observed in the manner of 5 Working Example 1. As a result, no void was observed in any of the ceramic green chips.

[0204]

Working Example 7

A dielectric paste for forming a spacer layer was prepared in the 10 manner of Working Example 5 except that ethyl cellulose having a weight average molecular weight of 130,000 and ethyl cellulose having a weight average molecular weight of 230,000 at a weight ratio of 75:25, namely, ethyl cellulose having an apparent weight average molecular weight of 155,000, was used as a binder of the dielectric paste and the viscosity of 15 the thus prepared conductive paste was measured under conditions of a temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also measured under conditions of a temperature of 25 °C and shearing velocity of 50 sec⁻¹.

[0205]

20 As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec⁻¹ was 14.9 Ps·s and that the viscosity of the dielectric paste measured under condition of the shearing velocity of 50 sec⁻¹ was 8.77 Ps·s.

[0206]

25 Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner of Working Example 1, thereby forming a spacer layer.

[0207]

Further, the surface of the thus formed spacer layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the spacer layer was free of cracks and wrinkles.

5 [0208]

A conductive paste for forming an electrode layer was then prepared in the manner of Working Example 1 except that dihydroterpinyl methyl ether was used as a solvent instead of isobornyl acetate and the thus prepared conductive paste was printed on the 10 ceramic green sheet, thereby fabricating a multi-layered unit including the electrode layer and the spacer layer laminated on the ceramic green sheet.

[0209]

Further, the surface of the thus formed electrode layer was 15 observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the electrode layer was free of cracks and wrinkles.

[0210]

Furthermore, a total of thirty annealing treated ceramic green 20 chips were fabricated in the manner of Working Example 1 and the surface of each of the ceramic green chips was observed in the manner of Working Example 1. As a result, no void was observed in any of the ceramic green chips.

[0211]

25 Working Example 8

A dielectric paste for forming a spacer layer was prepared in the manner of Working Example 5 except that ethyl cellulose having a weight average molecular weight of 130,000 and ethyl cellulose having a weight

average molecular weight of 230,000 at a weight ratio of 50:50, namely, ethyl cellulose having an apparent weight average molecular weight of 180,000, was used as a binder of the dielectric paste and the viscosity of the thus prepared conductive paste was measured under conditions of a 5 temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also measured under conditions of a temperature of 25 °C and shearing velocity of 50 sec⁻¹.

[0212]

As a result, it was found that the viscosity of the dielectric paste 10 measured under condition of the shearing velocity of 8 sec⁻¹ was 19.0 Ps·s and that the viscosity of the dielectric paste measured under condition of the shearing velocity of 50 sec⁻¹ was 11.2 Ps·s.

[0213]

Then, the thus prepared dielectric paste was printed using a 15 screen printing machine on a ceramic green sheet formed in the manner of Working Example 1, thereby forming a spacer layer.

[0214]

Further, the surface of the thus formed spacer layer was observed 20 at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the spacer layer was free of cracks and wrinkles.

[0215]

A conductive paste for forming an electrode layer was then prepared in the manner of Working Example 1 except that 25 dihydroterpinyl methyl ether was used as a solvent instead of isobornyl acetate and the thus prepared conductive paste was printed on the ceramic green sheet, thereby fabricating a multi-layered unit including the electrode layer and the spacer layer laminated on the ceramic green

sheet.

[0216]

Further, the surface of the thus formed electrode layer was observed at four-hundred magnifications using a metallographic 5 microscope. As a result, it was found that the surface of the electrode layer was free of cracks and wrinkles.

[0217]

Furthermore, a total of thirty annealing treated ceramic green chips were fabricated in the manner of Working Example 1 and the 10 surface of each of the ceramic green chips was observed in the manner of Working Example 1. As a result, no void was observed in any of the ceramic green chips.

[0218]

Comparative Example 5

15 A dielectric paste for forming a spacer layer was prepared in the manner of Working Example 5 except that ethyl cellulose having a weight average molecular weight of 75,000 and ethyl cellulose having a weight average molecular weight of 130,000 at a weight ratio of 50:50, namely, ethyl cellulose having an apparent weight average molecular weight of 20 102,500, was used as a binder of the dielectric paste and the viscosity of the thus prepared conductive paste was measured under conditions of a temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also measured under conditions of a temperature of 25 °C and shearing velocity of 50 sec⁻¹.

25 [0219]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec⁻¹ was 4.30 Ps·s and that the viscosity of the dielectric paste measured under condition of

the shearing velocity of 50 sec^{-1} was $3.10\text{ Ps}\cdot\text{s}$.

[0220]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner of Working Example 1. However, since the viscosity of the dielectric paste was too low, a spacer layer could not be formed.

[0221]

Comparative Example 6

A dielectric paste for forming a spacer layer was prepared in the manner of Working Example 5 except that ethyl cellulose having a weight average molecular weight of 130,000 and ethyl cellulose having a weight average molecular weight of 230,000 at a weight ratio of 25:75, namely, ethyl cellulose having an apparent weight average molecular weight of 205,000, was used as a binder of the dielectric paste and the viscosity of the thus prepared conductive paste was measured under conditions of a temperature of $25\text{ }^{\circ}\text{C}$ and shearing velocity of 8 sec^{-1} and was also measured under conditions of a temperature of $25\text{ }^{\circ}\text{C}$ and shearing velocity of 50 sec^{-1} .

[0222]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec^{-1} was $23.9\text{ Ps}\cdot\text{s}$ and that the viscosity of the dielectric paste measured under condition of the shearing velocity of 50 sec^{-1} was $14.0\text{ Ps}\cdot\text{s}$.

[0223]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner of Working Example 1. However, since the viscosity of the dielectric paste was too high, the clogging of a screen printing plate occurred and a

continuous spacer layer could not be formed.

[0224]

Comparative Example 7

A dielectric paste for forming a spacer layer was prepared in the
5 manner of Working Example 5 except that ethyl cellulose having a weight
average molecular weight of 230,000 was used as a binder of the dielectric
paste and the viscosity of the thus prepared conductive paste was
measured under conditions of a temperature of 25 °C and shearing
velocity of 8 sec⁻¹ and was also measured under conditions of a
10 temperature of 25 °C and shearing velocity of 50 sec⁻¹.

[0225]

As a result, it was found that the viscosity of the dielectric paste
measured under condition of the shearing velocity of 8 sec⁻¹ was 32.2 Ps·s
and that the viscosity of the dielectric paste measured under condition of
15 the shearing velocity of 50 sec⁻¹ was 18.8 Ps·s.

[0226]

Then, the thus prepared dielectric paste was printed using a
screen printing machine on a ceramic green sheet formed in the manner
of Working Example 1. However, since the viscosity of the dielectric paste
20 was too high, the clogging of a screen printing plate occurred and a
continuous spacer layer could not be formed.

[0227]

Comparative Example 8

A dielectric paste for forming a ceramic green sheet was prepared
25 in the manner of Working Example 1 except that butyral system resin
whose degree of polymerization was 800 and degree of butyralization was
69 mol % was used as a binder of the dielectric paste for forming a
ceramic green sheet, thereby forming a ceramic green sheet.

Further, the thus prepared dielectric paste was printed using a screen printing machine in the manner of Working Example 81 on a ceramic green sheet formed in the manner of Working Example 1, thereby forming a spacer layer.

5 Further, the surface of the thus formed spacer layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that cracks and wrinkles were generated on the surface of the spacer layer.

[0228]

10 A conductive paste for forming an electrode layer was then prepared in the manner of Working Example 1 and the thus prepared conductive paste was printed on the ceramic green sheet, thereby fabricating a multi-layered unit including the electrode layer and the spacer layer laminated on the ceramic green sheet.

15 [0229]

Further, the surface of the thus formed electrode layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that cracks and wrinkles were generated on the surface of the electrode layer.

20 [0230]

Furthermore, a total of thirty annealing treated ceramic green chips were fabricated in the manner of Working Example 1 and the surface of each of the ceramic green chips was observed. As a result, voids were observed in the two ceramic green chips among the thirty ceramic green chips.

25 [0231]

Working Example 9

A dielectric paste was prepared in the manner of Working Example

1 except that terpinyl methyl ether was used as a solvent instead of isobornyl acetate and the viscosity of the thus prepared dielectric paste was measured under conditions of a temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also measured under conditions of a 5 temperature of 25 °C and shearing velocity of 50 sec⁻¹.

[0232]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec⁻¹ was 7.51 Ps·s and that the viscosity of the dielectric paste measured under condition of 10 the shearing velocity of 50 sec⁻¹ was 4.38 Ps·s.

[0233]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner of Working Example 1, thereby forming a spacer layer.

15 [0234]

Further, the surface of the thus formed spacer layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the spacer layer was free of cracks and wrinkles.

20 [0235]

A conductive paste for forming an electrode layer was then prepared in the manner of Working Example 1 except that terpinyl methyl ether was used as a solvent instead of isobornyl acetate and the thus prepared conductive paste was printed on the ceramic green sheet, 25 thereby fabricating a multi-layered unit including the electrode layer and the spacer layer laminated on the ceramic green sheet.

[0236]

Further, the surface of the thus formed electrode layer was

observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the electrode layer was free of cracks and wrinkles.

[0237]

5 Furthermore, a total of thirty annealing treated ceramic green chips were fabricated in the manner of Working Example 1 and the surface of each of the ceramic green chips was observed in the manner of Working Example 1. As a result, no void was observed in any of the ceramic green chips.

10 [0238]

Working Example 10

A dielectric paste for forming a spacer layer was prepared in the manner of Working Example 9 except that ethyl cellulose having a weight average molecular weight of 130,000 was used as a binder of the dielectric 15 paste and the viscosity of the thus prepared conductive paste was measured under conditions of a temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also measured under conditions of a temperature of 25 °C and shearing velocity of 50 sec⁻¹.

[0239]

20 As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec⁻¹ was 10.6 Ps·s and that the viscosity of the dielectric paste measured under condition of the shearing velocity of 50 sec⁻¹ was 6.34 Ps·s.

[0240]

25 Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner of Working Example 1, thereby forming a spacer layer.

[0241]

Further, the surface of the thus formed spacer layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the spacer layer was free of cracks and wrinkles.

5 [0242]

A conductive paste for forming an electrode layer was then prepared in the manner of Working Example 1 except that terpinyl methyl ether was used as a solvent instead of isobornyl acetate and the thus prepared conductive paste was printed on the ceramic green sheet, 10 thereby fabricating a multi-layered unit including the electrode layer and the spacer layer laminated on the ceramic green sheet.

[0243]

Further, the surface of the thus formed electrode layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the electrode layer was free of cracks and wrinkles.

[0244]

Furthermore, a total of thirty annealing treated ceramic green chips were fabricated in the manner of Working Example 1 and the 20 surface of each of the ceramic green chips was observed in the manner of Working Example 1. As a result, no void was observed in any of the ceramic green chips.

[0245]

Working Example 11

25 A dielectric paste for forming a spacer layer was prepared in the manner of Working Example 9 except that ethyl cellulose having a weight average molecular weight of 130,000 and ethyl cellulose having a weight average molecular weight of 230,000 at a weight ratio of 75:25, namely,

ethyl cellulose having an apparent weight average molecular weight of 155,000, was used as a binder of the dielectric paste and the viscosity of the thus prepared conductive paste was measured under conditions of a temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also 5 measured under conditions of a temperature of 25 °C and shearing velocity of 50 sec⁻¹.

[0246]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec⁻¹ was 14.7 Ps·s 10 and that the viscosity of the dielectric paste measured under condition of the shearing velocity of 50 sec⁻¹ was 8.56 Ps·s.

[0247]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner 15 of Working Example 1, thereby forming a spacer layer.

[0248]

Further, the surface of the thus formed spacer layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the spacer layer was free of cracks 20 and wrinkles.

[0249]

A conductive paste for forming an electrode layer was then prepared in the manner of Working Example 1 except that terpinyl methyl ether was used as a solvent instead of isobornyl acetate and the 25 thus prepared conductive paste was printed on the ceramic green sheet, thereby fabricating a multi-layered unit including the electrode layer and the spacer layer laminated on the ceramic green sheet.

[0250]

Further, the surface of the thus formed electrode layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the electrode layer was free of cracks and wrinkles.

5 [0251]

Furthermore, a total of thirty annealing treated ceramic green chips were fabricated in the manner of Working Example 1 and the surface of each of the ceramic green chips was observed in the manner of Working Example 1. As a result, no void was observed in any of the 10 ceramic green chips.

[0252]

Working Example 12

A dielectric paste for forming a spacer layer was prepared in the manner of Working Example 9 except that ethyl cellulose having a weight 15 average molecular weight of 130,000 and ethyl cellulose having a weight average molecular weight of 230,000 at a weight ratio of 50:50, namely, ethyl cellulose having an apparent weight average molecular weight of 180,000, was used as a binder of the dielectric paste and the viscosity of the thus prepared conductive paste was measured under conditions of a 20 temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also measured under conditions of a temperature of 25 °C and shearing velocity of 50 sec⁻¹.

[0253]

As a result, it was found that the viscosity of the dielectric paste 25 measured under condition of the shearing velocity of 8 sec⁻¹ was 18.8 Ps·s and that the viscosity of the dielectric paste measured under condition of the shearing velocity of 50 sec⁻¹ was 10.9 Ps·s.

[0254]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner of Working Example 1, thereby forming a spacer layer.

[0255]

5 Further, the surface of the thus formed spacer layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the spacer layer was free of cracks and wrinkles.

[0256]

10 A conductive paste for forming an electrode layer was then prepared in the manner of Working Example 1 except that terpinyl methyl ether was used as a solvent instead of isobornyl acetate and the thus prepared conductive paste was printed on the ceramic green sheet, thereby fabricating a multi-layered unit including the electrode layer and 15 the spacer layer laminated on the ceramic green sheet.

[0257]

Further, the surface of the thus formed electrode layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the electrode 20 layer was free of cracks and wrinkles.

[0258]

Furthermore, a total of thirty annealing treated ceramic green chips were fabricated in the manner of Working Example 1 and the surface of each of the ceramic green chips was observed in the manner of 25 Working Example 1. As a result, no void was observed in any of the ceramic green chips.

[0259]

Comparative Example 9

A dielectric paste for forming a spacer layer was prepared in the manner of Working Example 9 except that ethyl cellulose having a weight average molecular weight of 75,000 and ethyl cellulose having a weight average molecular weight of 130,000 at a weight ratio of 50:50, namely,
5 ethyl cellulose having an apparent weight average molecular weight of 102,500, was used as a binder of the dielectric paste and the viscosity of the thus prepared conductive paste was measured under conditions of a temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also measured under conditions of a temperature of 25 °C and shearing
10 velocity of 50 sec⁻¹.

[0260]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec⁻¹ was 4.22 Ps·s and that the viscosity of the dielectric paste measured under condition of
15 the shearing velocity of 50 sec⁻¹ was 2.91 Ps·s.

[0261]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner of Working Example 1. However, since the viscosity of the dielectric paste
20 was too low, a spacer layer could not be formed.

[0262]

Comparative Example 10

A dielectric paste for forming a spacer layer was prepared in the manner of Working Example 9 except that ethyl cellulose having a weight average molecular weight of 130,000 and ethyl cellulose having a weight average molecular weight of 230,000 at a weight ratio of 25:75, namely,
25 ethyl cellulose having an apparent weight average molecular weight of 205,000, was used as a binder of the dielectric paste and the viscosity of

the thus prepared conductive paste was measured under conditions of a temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also measured under conditions of a temperature of 25 °C and shearing velocity of 50 sec⁻¹.

5 [0263]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec⁻¹ was 24.2 Ps·s and that the viscosity of the dielectric paste measured under condition of the shearing velocity of 50 sec⁻¹ was 13.7 Ps·s.

10 [0264]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner of Working Example 1. However, since the viscosity of the dielectric paste was too high, the clogging of a screen printing plate occurred and a 15 continuous spacer layer could not be formed.

[0265]

Comparative Example 11

A dielectric paste for forming a spacer layer was prepared in the manner of Working Example 9 except that ethyl cellulose having a weight 20 average molecular weight of 230,000 was used as a binder of the dielectric paste and the viscosity of the thus prepared conductive paste was measured under conditions of a temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also measured under conditions of a temperature of 25 °C and shearing velocity of 50 sec⁻¹.

25 [0266]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec⁻¹ was 32.0 Ps·s and that the viscosity of the dielectric paste measured under condition of

the shearing velocity of 50 sec⁻¹ was 18.7 Ps·s.

[0267]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner of Working Example 1. However, since the viscosity of the dielectric paste was too high, the clogging of a screen printing plate occurred and a continuous spacer layer could not be formed.

[0268]

Comparative Example 12

A dielectric paste for forming a ceramic green sheet was prepared in the manner of Working Example 1 except that butyral system resin whose degree of polymerization was 800 and degree of butyralization was 69 mol % was used as a binder of the dielectric paste for forming a ceramic green sheet, thereby forming a ceramic green sheet.

Further, the thus prepared dielectric paste was printed using a screen printing machine in the manner of Working Example 12 on a ceramic green sheet formed in the manner of Working Example 1, thereby forming a spacer layer.

Further, the surface of the thus formed spacer layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that cracks and wrinkles were generated on the surface of the spacer layer.

[0269]

A conductive paste for forming an electrode layer was then prepared in the manner of Working Example 1 except that terpinyl methyl ether was used as a solvent instead of isobornyl acetate and the thus prepared conductive paste was printed on the ceramic green sheet, thereby fabricating a multi-layered unit including the electrode layer and

the spacer layer laminated on the ceramic green sheet.

[0270]

Further, the surface of the thus formed electrode layer was observed at four-hundred magnifications using a metallographic 5 microscope. As a result, it was found that cracks and wrinkles were generated on the surface of the electrode layer.

[0271]

Furthermore, a total of thirty annealing treated ceramic green 10 chips were fabricated in the manner of Working Example 1 and, the surface of each of the ceramic green chips was observed. As a result, voids were observed in the two ceramic green chips among the thirty ceramic green chips.

[0272]

Working Example 13

15 A dielectric paste was prepared in the manner of Working Example 2 except that α -terpinyl acetate was used as a solvent instead of isobornyl acetate and the viscosity of the thus prepared dielectric paste was measured under conditions of a temperature of 25 °C and shearing velocity of 8 sec^{-1} and was also measured under conditions of a 20 temperature of 25 °C and shearing velocity of 50 sec^{-1} .

[0273]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec^{-1} was 11.2 $\text{Ps}\cdot\text{s}$ and that the viscosity of the dielectric paste measured under condition of 25 the shearing velocity of 50 sec^{-1} was 5.69 $\text{Ps}\cdot\text{s}$.

[0274]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner

of Working Example 1, thereby forming a spacer layer.

[0275]

Further, the surface of the thus formed spacer layer was observed at four-hundred magnifications using a metallographic microscope. As a 5 result, it was found that the surface of the spacer layer was free of cracks and wrinkles.

[0276]

A conductive paste for forming an electrode layer was then prepared in the manner of Working Example 1 except that terpinyl 10 oxyethanol was used as a solvent instead of isobornyl acetate and the thus prepared conductive paste was printed on the ceramic green sheet, thereby fabricating a multi-layered unit including the electrode layer and the spacer layer laminated on the ceramic green sheet.

[0277]

15 Further, the surface of the thus formed electrode layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the electrode layer was free of cracks and wrinkles.

[0278]

20 Furthermore, a total of thirty annealing treated ceramic green chips were fabricated in the manner of Working Example 1 and the surface of each of the ceramic green chips was observed in the manner of Working Example 1. As a result, no void was observed in any of the ceramic green chips.

25 [0279]

Working Example 14

A dielectric paste was prepared in the manner of Working Example 2 except that I-dihydrocarvyl acetate was used as a solvent instead of

isobornyl acetate and the viscosity of the thus prepared dielectric paste was measured under conditions of a temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also measured under conditions of a temperature of 25 °C and shearing velocity of 50 sec⁻¹.

5 [0280]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec⁻¹ was 10.8 Ps·s and that the viscosity of the dielectric paste measured under condition of the shearing velocity of 50 sec⁻¹ was 6.62 Ps·s.

10 [0281]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner of Working Example 1, thereby forming a spacer layer.

[0282]

15 Further, the surface of the thus formed spacer layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the spacer layer was free of cracks and wrinkles.

[0283]

20 A conductive paste for forming an electrode layer was then prepared in the manner of Working Example 1 except that 1-dihydrocarvyl acetate was used as a solvent instead of isobornyl acetate and the thus prepared conductive paste was printed on the ceramic green sheet, thereby fabricating a multi-layered unit including the electrode 25 layer and the spacer layer laminated on the ceramic green sheet.

[0284]

Further, the surface of the thus formed electrode layer was observed at four-hundred magnifications using a metallographic

microscope. As a result, it was found that the surface of the electrode layer was free of cracks and wrinkles.

[0285]

Furthermore, a total of thirty annealing treated ceramic green 5 chips were fabricated in the manner of Working Example 1 and the surface of each of the ceramic green chips was observed in the manner of Working Example 1. As a result, no void was observed in any of the ceramic green chips.

[0286]

10 Working Example 15

A dielectric paste was prepared in the manner of Working Example 2 except that I-menthyl acetate was used as a solvent instead of isobornyl acetate and the viscosity of the thus prepared dielectric paste was measured under conditions of a temperature of 25 °C and shearing 15 velocity of 8 sec⁻¹ and was also measured under conditions of a temperature of 25 °C and shearing velocity of 50 sec⁻¹.

[0287]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec⁻¹ was 9.95 Ps·s 20 and that the viscosity of the dielectric paste measured under condition of the shearing velocity of 50 sec⁻¹ was 5.59 Ps·s.

[0288]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner 25 of Working Example 1, thereby forming a spacer layer.

[0289]

Further, the surface of the thus formed spacer layer was observed at four-hundred magnifications using a metallographic microscope. As a

result, it was found that the surface of the spacer layer was free of cracks and wrinkles.

[0290]

A conductive paste for forming an electrode layer was then
5 prepared in the manner of Working Example 1 except that I-menthyl acetate was used as a solvent instead of isobornyl acetate and the thus prepared conductive paste was printed on the ceramic green sheet, thereby fabricating a multi-layered unit including the electrode layer and the spacer layer laminated on the ceramic green sheet.

10 [0291]

Further, the surface of the thus formed electrode layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the electrode layer was free of cracks and wrinkles.

15 [0292]

Furthermore, a total of thirty annealing treated ceramic green chips were fabricated in the manner of Working Example 1 and the surface of each of the ceramic green chips was observed in the manner of Working Example 1. As a result, no void was observed in any of the
20 ceramic green chips.

[0293]

Working Example 16

A dielectric paste was prepared in the manner of Working Example 2 except that I-menthone was used as a solvent instead of isobornyl acetate and the viscosity of the thus prepared dielectric paste was measured under conditions of a temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also measured under conditions of a temperature of 25 °C and shearing velocity of 50 sec⁻¹.

[0294]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec^{-1} was $11.6 \text{ Ps}\cdot\text{s}$ and that the viscosity of the dielectric paste measured under condition of 5 the shearing velocity of 50 sec^{-1} was $6.43 \text{ Ps}\cdot\text{s}$.

[0295]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner of Working Example 1, thereby forming a spacer layer.

10 [0296]

Further, the surface of the thus formed spacer layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the spacer layer was free of cracks and wrinkles.

15 [0297]

A conductive paste for forming an electrode layer was then prepared in the manner of Working Example 1 except that I-menthone was used as a solvent instead of isobornyl acetate and the thus prepared conductive paste was printed on the ceramic green sheet, thereby 20 fabricating a multi-layered unit including the electrode layer and the spacer layer laminated on the ceramic green sheet.

[0298]

Further, the surface of the thus formed electrode layer was observed at four-hundred magnifications using a metallographic 25 microscope. As a result, it was found that the surface of the electrode layer was free of cracks and wrinkles.

[0299]

Furthermore, a total of thirty annealing treated ceramic green

chips were fabricated in the manner of Working Example 1 and the surface of each of the ceramic green chips was observed in the manner of Working Example 1. As a result, no void was observed in any of the ceramic green chips.

5 [0300]

Working Example 17

A dielectric paste was prepared in the manner of Working Example 2 except that I-perillyl acetate was used as a solvent instead of isobornyl acetate and the viscosity of the thus prepared dielectric paste was measured under conditions of a temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also measured under conditions of a temperature of 25 °C and shearing velocity of 50 sec⁻¹.

10 [0301]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec⁻¹ was 11.0 Ps·s and that the viscosity of the dielectric paste measured under condition of the shearing velocity of 50 sec⁻¹ was 5.87 Ps·s.

15 [0302]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner of Working Example 1, thereby forming a spacer layer.

20 [0303]

Further, the surface of the thus formed spacer layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the spacer layer was free of cracks and wrinkles.

25 [0304]

A conductive paste for forming an electrode layer was then

prepared in the manner of Working Example 1 except that I-perillyl acetate was used as a solvent instead of isobornyl acetate and the thus prepared conductive paste was printed on the ceramic green sheet, thereby fabricating a multi-layered unit including the electrode layer and 5 the spacer layer laminated on the ceramic green sheet.

[0305]

Further, the surface of the thus formed electrode layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the electrode 10 layer was free of cracks and wrinkles.

[0306]

Furthermore, a total of thirty annealing treated ceramic green chips were fabricated in the manner of Working Example 1 and the surface of each of the ceramic green chips was observed in the manner of 15 Working Example 1. As a result, no void was observed in any of the ceramic green chips.

[0307]

Working Example 18

A dielectric paste was prepared in the manner of Working Example 20 2 except that I-carvyl acetate was used as a solvent instead of isobornyl acetate and the viscosity of the thus prepared dielectric paste was measured under conditions of a temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also measured under conditions of a temperature of 25 °C and shearing velocity of 50 sec⁻¹.

25 [0308]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec⁻¹ was 10.2 Ps·s and that the viscosity of the dielectric paste measured under condition of

the shearing velocity of 50 sec⁻¹ was 5.69 Ps·s.

[0309]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner 5 of Working Example 1, thereby forming a spacer layer.

[0310]

Further, the surface of the thus formed spacer layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the spacer layer was free of cracks 10 and wrinkles.

[0311]

A conductive paste for forming an electrode layer was then prepared in the manner of Working Example 1 except that I-carvyl acetate was used as a solvent instead of isobornyl acetate and the thus 15 prepared conductive paste was printed on the ceramic green sheet, thereby fabricating a multi-layered unit including the electrode layer and the spacer layer laminated on the ceramic green sheet.

[0312]

Further, the surface of the thus formed electrode layer was 20 observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the electrode layer was free of cracks and wrinkles.

[0313]

Furthermore, a total of thirty annealing treated ceramic green 25 chips were fabricated in the manner of Working Example 1 and the surface of each of the ceramic green chips was observed in the manner of Working Example 1. As a result, no void was observed in any of the ceramic green chips.

[0314]

Comparative Example 13

A dielectric paste was prepared in the manner of Working Example 2 except that a mixed solvent of terpineol and kerosene (mixture ratio 5 (mass ratio) of 50:50) was used as a solvent instead of isobornyl acetate and the viscosity of the thus prepared dielectric paste was measured under conditions of a temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also measured under conditions of a temperature of 25 °C and shearing velocity of 50 sec⁻¹.

10 [0315]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec⁻¹ was 10.0 Ps·s and that the viscosity of the dielectric paste measured under condition of the shearing velocity of 50 sec⁻¹ was 6.43 Ps·s.

15 [0316]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner of Working Example 1, thereby forming a spacer layer.

[0317]

20 Further, the surface of the thus formed spacer layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that cracks and wrinkles were generated on the surface of the spacer layer.

[0318]

25 A conductive paste for forming an electrode layer was then prepared in the manner of Working Example 1 and the thus prepared conductive paste was printed on the ceramic green sheet, thereby fabricating a multi-layered unit including the electrode layer and the

spacer layer laminated on the ceramic green sheet.

[0319]

Further, the surface of the thus formed electrode layer was observed at four-hundred magnifications using a metallographic 5 microscope. As a result, it was found that the surface of the electrode layer was free of cracks and wrinkles.

[0320]

Furthermore, a total of thirty annealing treated ceramic green 10 chips were fabricated in the manner of Working Example 1 and the surface of each of the ceramic green chips was observed. As a result, voids 15 were observed in the eight ceramic green chips among the thirty ceramic green chips.

[0321]

Comparative Example 14

15 A dielectric paste was prepared in the manner of Working Example 2 except that terpineol was used as a solvent instead of isobornyl acetate and the viscosity of the thus prepared dielectric paste was measured under conditions of a temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also measured under conditions of a temperature of 25 °C and 20 shearing velocity of 50 sec⁻¹.

[0322]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec⁻¹ was 12.2 Ps·s and that the viscosity of the dielectric paste measured under condition of 25 the shearing velocity of 50 sec⁻¹ was 6.62 Ps·s.

[0323]

Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner

of Working Example 1, thereby forming a spacer layer.

[0324]

Further, the surface of the thus formed spacer layer was observed at four-hundred magnifications using a metallographic microscope. As a 5 result, it was found that cracks and wrinkles were generated on the surface of the spacer layer.

[0325]

A conductive paste for forming an electrode layer was then prepared in the manner of Working Example 1 and the thus prepared 10 conductive paste was printed on the ceramic green sheet, thereby fabricating a multi-layered unit including the electrode layer and the spacer layer laminated on the ceramic green sheet.

[0326]

Further, the surface of the thus formed electrode layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the electrode 15 layer was free of cracks and wrinkles.

[0327]

Furthermore, a total of thirty annealing treated ceramic green 20 chips were fabricated in the manner of Working Example 1 and the surface of each of the ceramic green chips was observed. As a result, voids were observed in the fifteen ceramic green chips among the thirty ceramic green chips.

[0328]

25 Comparative Example 15

A dielectric paste was prepared in the manner of Working Example 2 except that butyl carbitol acetate was used as a solvent instead of isobornyl acetate and the viscosity of the thus prepared dielectric paste

was measured under conditions of a temperature of 25 °C and shearing velocity of 8 sec⁻¹ and was also measured under conditions of a temperature of 25 °C and shearing velocity of 50 sec⁻¹.

[0329]

5 As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec⁻¹ was 5.12 Ps·s and that the viscosity of the dielectric paste measured under condition of the shearing velocity of 50 sec⁻¹ was 3.36 Ps·s.

[0330]

10 Then, the thus prepared dielectric paste was printed using a screen printing machine on a ceramic green sheet formed in the manner of Working Example 1. However, since the viscosity of the dielectric paste was too low, a spacer layer could not be formed.

[0331]

15 Comparative Example 16

A dielectric paste was prepared in the manner of Working Example 2 except that dihydroterpineol was used as a solvent instead of isobornyl acetate and the viscosity of the thus prepared dielectric paste was measured under conditions of a temperature of 25 °C and shearing 20 velocity of 8 sec⁻¹ and was also measured under conditions of a temperature of 25 °C and shearing velocity of 50 sec⁻¹.

[0332]

As a result, it was found that the viscosity of the dielectric paste measured under condition of the shearing velocity of 8 sec⁻¹ was 12.5 Ps·s 25 and that the viscosity of the dielectric paste measured under condition of the shearing velocity of 50 sec⁻¹ was 6.52 Ps·s.

[0333]

Then, the thus prepared dielectric paste was printed using a

screen printing machine on a ceramic green sheet formed in the manner of Working Example 1, thereby forming a spacer layer.

[0334]

Further, the surface of the thus formed spacer layer was observed 5 at four-hundred magnifications using a metallographic microscope. As a result, it was found that cracks and wrinkles were generated on the surface of the spacer layer.

[0335]

A conductive paste for forming an electrode layer was then 10 prepared in the manner of Working Example 1 and the thus prepared conductive paste was printed on the ceramic green sheet, thereby fabricating a multi-layered unit including the electrode layer and the spacer layer laminated on the ceramic green sheet.

[0336]

15 Further, the surface of the thus formed electrode layer was observed at four-hundred magnifications using a metallographic microscope. As a result, it was found that the surface of the electrode layer was free of cracks and wrinkles.

[0337]

20 Furthermore, a total of thirty annealing treated ceramic green chips were fabricated in the manner of Working Example 1 and the surface of each of the ceramic green chips was observed. As a result, voids were observed in the nine ceramic green chips among the thirty ceramic green chips.

25 [0338]

It was found from Working Examples 1 to 18 and Comparative Examples 13 to 16 that in the case where a dielectric paste adapted for forming a spacer layer and containing ethyl cellulose having a weight

average molecular weight of 130,000 as a binder and the mixed solvent of terpineol and kerosene (mixture ratio (mass ratio) of 50:50) as a solvent, a dielectric paste adapted for forming a spacer layer and containing ethyl cellulose having a weight average molecular weight of 130,000 as a binder
5 and terpineol as a solvent, a dielectric paste adapted for forming a spacer layer and containing ethyl cellulose having a weight average molecular weight of 130,000 as a binder and butyl carbitol acetate as a solvent or a dielectric paste adapted for forming a spacer layer and containing ethyl cellulose having a weight average molecular weight of 130,000 as a binder
10 and dihydroterpineol as a solvent was printed on the ceramic green sheet formed by using the dielectric paste containing polyvinyl butyral whose degree of polymerization was 1450 and degree of butyralization was 69 mol % as a binder, thereby fabricating the multi-layered unit, and fifty of the multi-layered units were laminated, thereby fabricating the ceramic
15 green chip, a spacer layer itself could not be formed or even if a spacer layer could be formed, cracks and wrinkles were generated on the surface of the spacer layer and voids were generated in a ceramic green chip fabricated by laminating the multi-layered units to form a laminated body and baking the laminated body, while in the case where a dielectric paste
20 adapted for forming a spacer layer and containing ethyl cellulose having an apparent weight average molecular weight of 116,250 to 180,000 as a binder and isobornyl acetate, dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate or I-carvyl acetate as a solvent was
25 printed on the ceramic green sheet formed by using the dielectric paste containing polyvinyl butyral whose degree of polymerization was 1450 and degree of butyralization was 69 mol % as a binder, thereby fabricating the multi-layered unit, and fifty of the multi-layered units

were laminated, thereby fabricating the ceramic green chip, generation of cracks or wrinkles was not observed on the surface of the spacer layer and generation of voids was not observed in a ceramic green chip fabricated by laminating the multi-layered units to form a laminated body and baking 5 the laminated body.

[0339]

It is reasonable to conclude that this was because butyl carbitol acetate used as a solvent of the dielectric paste for forming a spacer layer in Comparative Example 15 does not dissolve polyvinyl butyral contained 10 as a binder in the dielectric paste used for forming the ceramic green sheet but the viscosity of the dielectric paste prepared is too low and because the mixed solvent of terpineol and kerosene (mixture ratio (mass ratio) of 50:50), terpineol and dihydroterpineol used as the solvent of the dielectric paste for forming the spacer layer in Comparative Examples 13, 15 14 and 16 dissolved polyvinyl butyral contained in the dielectric paste used for forming the ceramic green sheet and, therefore, the ceramic green sheet was swollen or partly dissolved, whereby voids were generated at the interface between the ceramic green sheet and the spacer layer or cracks and wrinkles were generated on the surface of the 20 spacer layer and voids were generated in the ceramic green chip fabricated by laminating the multi-layered units to form a laminated body and baking the laminated body or portions of the spacer layer where cracks and wrinkles were generated dropped off during the lamination of the multi-layered units, whereby voids were liable to be generated in the 25 ceramic green chip after baking, while isobornyl acetate, isobornyl acetate, dihydroterpinyl methyl ether, terpinyl methyl ether, α -terpinyl acetate, I-dihydrocarvyl acetate, I-menthyl acetate, I-menthone, I-perillyl acetate and I-carvyl acetate used as the solvent of the dielectric paste for forming

the spacer layer in Working Examples 1 to 18 hardly dissolved polyvinyl butyral contained in the dielectric paste used for forming the ceramic green sheet and it was possible to prevent cracks and wrinkles from being generated on the surface of the spacer layer and prevent voids from being 5 generated in the ceramic green chip after baking.

[0340]

Further, it was found from Working Examples 1 to 12, Comparative Examples 1, 5 and 9 and Comparative Examples 2, 3, 6, 7, 10 and 11 that in the case where even in the case where the dielectric 10 paste adapted for forming a spacer layer and containing isobornyl acetate, dihydroterpinyl methyl ether or terpinyl methyl ether as a solvent was printed on the ceramic green sheet formed by using the dielectric paste containing polyvinyl butyral whose degree of polymerization was 1450 and degree of butyralization was 69 mol % as a binder, thereby forming a 15 spacer layer, when ethyl cellulose having an apparent weight average molecular weight of 102,500 was used as a binder of the dielectric paste for forming a spacer layer, the viscosity of the dielectric paste for forming a spacer layer was so low that a spacer layer could not be formed and that on the other hand, even in the case where the dielectric paste adapted for 20 forming a spacer layer and containing isobornyl acetate, dihydroterpinyl methyl ether or terpinyl methyl ether as a solvent was printed on the ceramic green sheet formed by using the dielectric paste containing polyvinyl butyral whose degree of polymerization was 1450 and degree of butyralization was 69 mol % as a binder, thereby forming a spacer layer, 25 when ethyl cellulose having an apparent weight average molecular weight equal to or larger than 205,000 was used as a binder of the dielectric paste for forming a spacer layer, the viscosity of the dielectric paste for forming a spacer layer was so high that the clogging of a screen

printing plate occurred and a continuous spacer layer could not be formed and that it was preferable to use ethyl cellulose having an apparent weight average molecular weight larger than 102,500 and smaller than 205,000 as a binder of a dielectric paste for forming a spacer layer.

5 [0341]

Moreover, it was found from Working Examples 1 to 12 and Comparative Examples 4, 8 and 12 that even in the case where a dielectric paste adapted for forming a spacer layer and containing ethyl cellulose having an apparent weight average molecular weight larger than 102,500 and smaller than 205,000 as a binder and isobornyl acetate, dihydroterpinyl methyl ether or terpinyl methyl ether as a solvent was used to form a spacer layer, when a ceramic green sheet was formed by using a dielectric paste containing polyvinyl butyral whose degree of polymerization was 800 and degree of butyralization was 69 mol % as a binder, since a part of the binder of the dielectric paste for forming the ceramic green sheet was swollen or dissolved by the solvents contained in the dielectric paste used for forming the spacer layer and the conductive paste used for forming the electrode layer, voids were generated at the interface between the ceramic green sheet and the spacer layer and the electrode layer or cracks and wrinkles were generated on the surface of the spacer layer and the electrode layer and voids were generated in the ceramic green chip fabricated by laminating the multi-layered units to form a laminated body and baking the laminated body or portions of the spacer layer and the electrode layer where cracks and wrinkles were generated dropped off during the lamination of the multi-layered units, whereby voids were liable to be generated in the ceramic green chip after baking.

[0342]

The present invention has thus been shown and described with reference to the preferred embodiments and the working examples. However, it should be noted that the present invention is in no way limited to the details of the described arrangement but changes and 5 modifications may be made without departing from the scope of the appended claims.

[0343]

According to the present invention, it is possible to provide a dielectric paste for a spacer layer of a multi-layered ceramic electronic 10 component which does not dissolve a binder contained in a layer adjacent to the spacer layer of the multi-layered ceramic electronic component and can reliably prevent defects from being generated in a multi-layered ceramic electronic component.

[0344]

15 Further, according to the present invention, it is possible to provide a method for fabricating a multi-layered unit for a multi-layered ceramic electronic component which can reliably prevent defects from being generated in a multi-layered ceramic electronic component and form a spacer layer in a desired manner.

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